NONLINEAR PHASE QUASI-MATCHING AT THE THIRD HARMONIC GENERATION UNDER THE TWO-PHOTON RESONANCE

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The third harmonic generation (THG) under the conditions of a two-photon resonance is investigated for various nonlinear polarizabilities as a function of wave Δk and frequency $\Delta\omega$ mismatches from the resonance when taking into account the change of populations of resonant states. It is shown that, at a nonlinear phase quasi-matching (NPM), when the nonlinear wave mismatch at $\Delta\omega \neq 0$ is compensated by a linear one Δk, the efficiency of THG can increase approximately by 2 times in comparison with the case of $\Delta k = \Delta\omega = 0$ and reaches 60%. It is found that the fixed pump field approximation does not allow one to describe the NPM phenomenon, and it is necessary to use the fixed intensity approximation. It is shown analytically and numerically that, in the case of NPM, the wave and frequency mismatches can have both identical and different signs. In a spectrum of harmonics, a gap caused by backward Raman scattering can appear, which is getting deeper at large pump intensities due to the saturation of the populations of resonant states.

Introduction

The nonlinear mixing of frequencies is known to be one of the mostly developed methods for deriving the coherent radiation in a wide spectrum range [1—10]. Its effectiveness depends on the properties of nonlinear media and the intensities and divergence of the exciting beam. The use of the phase matching (PM) conditions [1, 2, 11] allows one to accumulate nonlinear effects with respect to the spatial coordinates during the propagation of waves and to significantly rise the effectiveness of the frequency transformation processes. Under non-resonance conditions, an effective transformation of a frequency requires the sufficiently high intensities of the exciting radiation. For this purpose, a high-level focusing of pumping radiation is used. At the sufficiently high intensities of the exciting radiation, an increase of the frequency transformation effectiveness is achieved by using the nonlinear parametric interaction of waves under the condition of the resonance between the frequencies of interacting waves and the energy states of the medium. In this case, the value of nonlinear susceptibility significantly increases. The resonance conditions can be also realized in crystalline media, but a resonance-induced increase of polarizabilities is inversely proportional to the energy width of the resonance states. That is why nonlinear resonance processes are more often dealt with in gas-like media (metal vapors, inert gases, etc.) having narrow energy levels [4—10, 12—16] and cryogenic liquids [3]. One can also employ the narrow energy levels of some rare-earth ions that are incorporated in crystals. In this case, the nonlinear susceptibilities increase by factor of $10^3 - 10^5$. One can use one-photon and two-photon resonances, as well as those of higher orders. In the resonance nonlinear optics, the most frequently are used two-photon resonances in the absence of the strong one-photon absorption at the frequencies of interacting waves [17, 18]. To create the sources of coherent radiation in the ultraviolet spectrum range, including vacuum ultraviolet and soft X-radiation range, the tripling of a laser radiation frequency is used, including the excimer lasers or tunable dye lasers, their harmonics, and sum frequencies [14—16, 19].

Here, we theoretically analyze the conditions for effective third harmonic generation in nonlinear media under the two-photon absorption resonance and the exciting laser radiation. Special attention is paid to a change of the PM conditions that arises due to significant variations of the dispersion of the medium in the resonance region induced by a high-intensity laser field.

From the physical point of view, resonance nonlinear optical processes are much more diversified than non-resonance ones [4—10]. In particular, this can be seen from the fact that, for the processes of parametric frequency transformation under resonance and quasi-resonance conditions, the relationship between the phases of interacting waves changes not only through their nonlinear interaction but also due to the resonance self-action accompanying the nonlinear processes [two-photon absorption (TPA) of pumping, backward Raman scattering (BRS), and generated radiation of the third harmonic] as well as a change of the populations of

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resonance states, multiphoton ionization, etc. These processes predetermine the conditions for a change of the refractive indices of the medium and the mismatch of wave vectors Δk, which is essential for the dynamics of nonlinear optical interaction [20,21]. Under these conditions, in addition to the linear wave mismatch due to the dispersion of the equilibrium medium, it is necessary to allow for the nonlinear wave mismatch Δk_{NL} depending on the changed dispersion, that is, on the intensities of interacting waves.

In order to describe the nonlinear optical processes under such conditions, we have introduced a concept of the NPM that assumes the compensation of a nonlinear wave mismatch with the linear one. The effectiveness of such a conception was demonstrated by the example of the generation a sum frequency under the TPA conditions [20,21]. The importance of analyzing the quasi-matching conditions is explained by the fact that the change of wave phases determines the energy exchange rate between interactive waves. In this case, the generalized phase characterizing the parametric interaction in the restricted spatial region of a nonlinear medium varies near some value which is optimal for the harmonic generation (a quasi-matching interaction). This allows one to improve the effectiveness of the transformation as against the case of linear quasi-matching without taking into account the induced dispersion of the medium.

In addition to the quasi-matching conditions in spatially homogeneous media, we also consider the phase matching of waves in specially fabricated heterogeneous solid-state lattice structures, where the phase frequently varies near the magnitude required for the phase transformation [1, 22]. Such a phase matching in the lattice structures is most often realized for nonresonance three-wave processes. A change of the NPM conditions under intensive pumping can be induced by a heat release. In particular, the generation of the second harmonic in crystal $ZnCs_2P_2$ can be accompanied by a change of the angles of matching by $5-25^{\circ}$ [23].

It is important that, in the case of resonance interactions of waves, the NPM under a slight deviation from a resonance can provide a significant increase of the effectiveness of the frequency transformation, which is impossible for nonresonance processes. It is explained by the fact that the effectiveness of resonance processes is essentially restricted by accompanying nonresonance processes, for example TAP, and the mismatch from the exact resonance decreases this negative influence. That's why the consideration of quasi-matching is closely connected to the problem of the greatest possible efficiency of the transformation. The specific feature of the GTH under the TPA conditions is the absence of a weak wave (like the case where the sum frequency is generated with the help of laser pumping) and the signal radiation, which requires a detailed theoretical analysis. In this case, it is important to allow for the essential role of the BRS of the generated radiation with amplification of the exciting radiation. It is worth mentioning that backward parametric processes under nonresonance interactions depend on phase conditions, while a part of backward processes (the incoherent ones) under resonance conditions does not depend on the generalized phase.

1. Problem Statement and Original Equations

In the present work, we investigate the NPM in case where the third harmonic $\omega_3 = 3\omega_1$ is generated under conditions of the TPR $2\omega_1 = \omega_{21} + \Delta\omega$. Here, ω_1 denotes the pump frequency, ω_{21} is the equivalent frequency of the dipole-forbidden transition between states 1 and 2 allowing a two-photon transition, and $\Delta\omega$ is the frequency mismatch from TPR. Let us consider the interaction of plane monochromatic waves propagating along the z axis and characterized with fixed linear polarizations: $\mathbf{E}_{1,3} = \mathbf{e}_{1,3}A_{1,3}\exp[i(\omega_{1,3}t - k_{1,3}z)] + \text{c.c.}$ specified by the orts of polarization $\mathbf{e}_{1,3}$.

The electromagnetic fields of the interacting waves $\omega_{1,3}$ are described by the system of Maxwell's equations interconnected through the nonlinear polarizations. The resonance nonlinear polarizations and the populations of resonance states $1, 2$ as well as the interaction of waves in the medium are considered in the course of time intervals much greater than the time of relaxation of the excited state population T_1, the time of polarization T_2, and that of transmission of radiation through the nonlinear medium. While dealing with the equations for the statistical matrix, we use the method of averaging over fast motions [4], while, in the wave equations due to small variations of the wave amplitudes at a distance in the order of a wavelength, we use the contraction procedure and retain only first-order derivatives with respect to the coordinate z, along which the propagation of waves and the accumulation of nonlinear interaction take place [1]. In this case, even with nonlinearities being respectively weak, the amplitude of the generated wave attains the values comparable to the amplitude of the exciting radiation.

In the considered quasistationary case, the solution of the problem is simplified significantly as it is easy to find analytic solutions for elements of the statistical matrix. Nonlinear processes under study are allowed for in the explicit form of the matrix element for the interaction of the fields with a generalized two-level system [4]. Moreover, the dynamics of resonance states is considered completely, while the whole set of other levels is allowed for the coefficients of the nonlinear polarizations describing the TPA of pumping ω_1 and stimulated Raman scattering $\omega_3 \to \omega_1 + \omega_{21} + \Delta\omega$. The solutions obtained for the statistical matrix allow one to find explicitly the nonlinear polarizations [4—10] contained in the coupled Maxwell's equations for interacting waves.

The system of truncated equations for the complex amplitudes $A_{1,3}$ for the process of third harmonic generation under the conditions of TPA with regard for the wave $\Delta k = k_3 - 3k_1$ and frequency $\Delta\omega = 2\omega_1 - \omega_{21}$ mismatchings looks as

$$\frac{dA_1}{dz} = -g_1 n[2(a-ib)\varkappa_1^2 A_1|A_1|^2 - (a+ib)\varkappa_2^2 \times$$
$$\times A_1|A_3|^2 + (a-3ib)\varkappa_1\varkappa_2 A_3 A_1^{*2}\exp(-i\Delta kz)],$$
$$\frac{dA_3}{dz} = -g_3 n(a-ib)(\varkappa_1\varkappa_2 A_1^3 \exp(i\Delta kz) + \varkappa_2^2 A_3|A_1|^2), \tag{1}$$

where we have introduced the notations

$$\Delta N = N_1 - N_2, \quad n = \frac{\Delta N}{|\Delta N_0|} = \mathrm{sign}(\Delta N_0)\times$$
$$\times\left\{1 + \frac{4aT_1T_2}{\hbar^2}\left[\varkappa_1^2|A_1|^4 + \varkappa_2^2|A_1A_3|^2 + \right.\right.$$
$$\left.\left. +\varkappa_1\varkappa_2\left(A_1^3A_3^*\exp(i\Delta kz) + \mathrm{h.c.}\right)\right]\right\}^{-1},$$

and

$$g_i = \frac{2\pi\omega_j^2 T_2|\Delta N_0|}{c^2\hbar k_j}, \quad a = 1/(1+\nu^2),$$
$$b = a\nu, \quad \nu = \Delta\omega T_2,$$
$$\varkappa_1 = \frac{2}{\hbar}\sum_k \frac{(\mathbf{d}_{1k}\mathbf{e}_1)(\mathbf{d}_{k2}\mathbf{e}_1)}{\omega_{k1}-\omega_1},$$
$$\varkappa_2 = \frac{1}{\hbar}\sum_k \frac{(\mathbf{d}_{1k}\mathbf{e}_3)(\mathbf{d}_{k2}\mathbf{e}_1)}{\omega_{k1}-\omega_3} + \frac{(\mathbf{d}_{1k}\mathbf{e}_1)(\mathbf{d}_{k2}\mathbf{e}_3)}{\omega_{k1}+\omega_1}.$$

Here, $N_{1,2}$ are the populations of levels 1, 2; ΔN — the absolute and n — the relative differences of the populations of the states, (ΔN_0 denotes the value of ΔN at $A_{1,3} = 0$); $\varkappa_{1,2}$ – the second-order polarizations describing the TAP at $\Delta N_0 > 0$ and the Stokes Raman scattering of the harmonic $\omega_3 \to \omega_1 + \omega_{21}$; $\mathbf{d}_{ik}$ are the matrix elements of the dipole moment of the transition between states i and k. In the expressions for the polarizations $\varkappa_{1,2}$, the summation is carried out over all levels k having the parity of the wave functions opposite to that of states 1, 2.

The first term of the right-hand term of Eq. (1) for the pump amplitude A_1 describes the two-photon absorption, while the second term in the both equations indicates the stimulated Raman scattering of a wave of the harmonic ω_3 resulting in the formation of the Stokes wave ω_1 with the participation of resonance states 1 and 2. The terms of Eq. (1) containing the phase multipliers $\exp(\Delta kr)$ describe the investigated process of GTH. The system of equations (1) allows for the complex character and the dispersion of nonlinear susceptibilities χ_R close to the resonance, whose form is taken to be Lorentz for the sake of simplicity. The dispersion of the real and imaginary parts of the nonlinear susceptibility is specified by the multipliers a and b. Thus, in Eq. (1), we allow for the accompanying two-photon processes as well as the variation of the populations of resonance states, which gives us a possibility to consider the noninverting $n > 0 (N_1 > N_2)$ and inverted $n < 0$ states of the medium. The process of self-action of the harmonic proportional to $\chi_{NR}A_3 \mid A_3 \mid^2$ is not taken into account in (1) due to the smallness of the nonresonance component of the nonlinear susceptibility χ_{NR}.

Let us introduce the expansible wave amplitudes, length and wave mismatch

$$a_j = \sqrt{\frac{g_1}{g_j}}\frac{\mid A_j \mid}{A_s}, \quad \xi = \frac{z}{l_0}, \quad s = \Delta k l_0;$$

$$l_0 = \left(g_1\varkappa_1^2 A_s^2\right)^{-1}, \ A_s = \sqrt{\frac{\hbar}{2\mid\varkappa_1\mid\sqrt{T_1T_2}}}, \tag{2}$$

where A_s denotes the saturating pump field, in which the initial difference between the populations of resonance states changes by a factor of two ($\Delta N(z=0) = \Delta N_0/2$) under the condition of the exact TPR ($\nu = 0$), and l_0 is the characteristic length of the TPR at $A_1 = A_s$. In the case of the TPR between the states $3s$ and $3d$ in a Na vapour at a pressure of 10 Torr ($\Delta N_0 = 1.17\cdot 10^{17}$ cm^{-3}), A_s corresponds to the intensity of 1.13 MW/cm^2 and $l_0 = 3.5$ cm [5].

Using (2), it is possible to pass from the equations for the complex amplitudes $A_{1,3}$ to those for the real ones $|A_j|$, and the generalized phase $\theta = s\xi + \varphi_3 - 3\varphi_1$ (here, φ_j are the phases of the complex amplitudes A_j of waves with the frequencies ω_j: $A_j = |A_j|\exp(-i\varphi_j)$). Such a representation appears to be more appropriate in many cases of a nonlinear interaction of waves, since it allows one to separate the values changing rapidly and slowly, decrease the number of variables, and simplify the derivation of both analytic and numerical solutions. Allowing for the dependence of the amplitude variation on a certain linear combination of the phases φ_j, it is possible to pass from four variables in the equations to three ones.

$$\frac{da_1}{d\xi} = -a_1\frac{n}{\varkappa}\left[a(2\varkappa a_1^2 - a_3^2/\varkappa + a_1 a_3\cos\theta) - \right.$$

$$\left. -3ba_1a_3\sin\theta\right],$$

$$\frac{da_3}{d\xi} = -a_1^2\frac{n}{\varkappa}\left[a(\frac{a_3}{\varkappa} + a_1\cos\theta) + ba_1\sin\theta\right],$$

$$\frac{d\theta}{d\xi} = s + a_1\frac{n}{\varkappa}\left\{a\left(3a_3 + \frac{a_1^2}{a_3}\right)\sin\theta + \right.$$

$$\left. +b\left[3\left(2\varkappa a_1 + \frac{a_3^2}{\varkappa a_1}\right) - \frac{a_1}{\varkappa} + \left(9a_3 - \frac{a_1^2}{a_3}\right)\cos\theta\right]\right\},$$

$$n = \left[1 + a\,a_1^2\left(a_1^2 + \frac{2a_1a_3}{\varkappa}\cos\theta + \frac{a_3^2}{\varkappa^2}\right)\right]^{-1},$$

$$\varkappa = \sqrt{\frac{g_1}{g_3}\frac{\varkappa_1}{\varkappa_2}}. \qquad (3)$$

If we use normalization (2), the GTH under the conditions of the TPR is characterized by the minimal number of independent dimensionless parameters [5—10]. In this case, the ratio of the squared amplitudes is equal to that of the densities of photon fluxes, and the effectiveness of the process can be characterized by the quantum conversion coefficient $\eta = a_3^2/a_{10}^2$, where a_{10} denotes the pump amplitude at $\xi = 0$.

2. Analytic Solutions in the Approximation of a Specified Intensity of Excitation

At the initial stage of GTH, the variation of $|A_1|$ at the expense of the TPA and the generation of a harmonic can be neglected, but the allowance should be made for a change of the phase $\varphi_1(\xi)$ due to the nonlinear dispersion in the quasiresonance region which is described by the real part of the corresponding cubic susceptibility. In this case, from the first equation of system (1), we deduce

$$a_1 = a_{10}\exp[i(2b\varkappa^2\zeta - \varphi_{10})], \qquad (4)$$

where

$$\zeta = \frac{z}{l_1} = \frac{n_0 a_{10}^2}{\varkappa^2}\xi,$$

$$l_1 = \left(g_3 n_0 \varkappa_2^2 |A_{10}|^2\right)^{-1}, \quad n_0 = \left(1 + a a_{10}^4\right)^{-1},$$

$$\varphi_{10} = \varphi_1(z=0).$$

In the approximation of a given intensity of pumping (4), the second equation of system (1) allows us to find the expressions for the amplitude of the harmonic and the quantum conversion coefficient

$$\eta = \frac{a_3^2}{a_{10}^2} = \varkappa^2\mathcal{F}(\zeta, a, \beta), \qquad (5)$$

where

$$\mathcal{F}(\zeta, a, \beta) = a\frac{[1 - e^{-a\zeta}]^2 + 4e^{-a\zeta}\sin^2(\beta\zeta/2)}{a^2 + \beta^2},$$

$$\beta = \sigma + \rho b, \quad \rho = 6\varkappa^2 - 1,$$

$$\sigma = \Delta k l_1 = \frac{\varkappa^2 s}{n_0 a_{10}^2}.$$

It is worth noting that the function $\mathcal{F}$ in the nonresonance case ($\nu \gg 1$, $a, b \ll 1$) turns into the known multiplier of PS $\mathcal{F}(\zeta, \sigma) = a\,\zeta^2\mathrm{sinc}^2(\sigma\zeta/2)$, where $\mathrm{sinc}(\tau) \equiv \sin\tau/\tau$ describing the known oscillations of the intensity of a generated radiation under the variation of the wave deviation Δk which appears in the expression for σ. These oscillations are discovered in the angular dependence of the generated radiation intensity as Maker strips but they can also manifest themselves under the variation of a pump wavelength. One of the resonance interaction differences of a wave from the nonresonance one lies in the non-zero intensity of the generated radiation at the minima of oscillations. At the same time, formula (4) describes the resonance nonlinear interaction of waves which is characterized by the wave antireflection of matter [5—10, 17]. In this case of a nonlinear medium, the fixed interconsistent amplitudes of all interacting waves, at which the absorption and the nonlinear interaction of waves disappear, are established, and a coherent superposition of waves propagates in the medium without damping.

In expression (5), the phenomenon of NPM is determined by the quantity β, where the term σ corresponds to a linear wave mismatch, while the term

$\rho b(\nu)$ defines a nonlinear one which depends, in turn, on the frequency mismatch ν.

Taking into account that the pump amplitude $A_1(z)$ at $|A_3| \ll |A_1|$ decreases mainly due to the TAP, let us find the domain of applicability of the approximation of a specified intensity

$$a\zeta a_{10}^2/(1+a_{10}^4) \ll 1. \quad (6)$$

If this criterion is violated, the analytic solution (5) describes the GTH only qualitatively.

3. The Nature of the Decreasing Effectiveness of the GTH in the Neighborhood of an Exact Resonance

Let us consider the quasiresonance GTH in the absence of a wave mismatch ($s = 0, \quad \nu \neq 0$). In this case, the expansion of the function $\mathcal{F}(\zeta, a, \beta)$ (5) in a series in terms of the frequency mismatch has a form

$$\mathcal{F} = \left(1 - e^{-\zeta}\right)^2 - \nu^2 f_1(\zeta, \rho) - \nu^4 f_2(\zeta, \rho), \quad (7)$$

where

$$f_1 = 2\left(\zeta - \rho^2\zeta^2/2\right)e^{-\zeta} - 2\zeta e^{-2\zeta} - (1-\rho^2)\left(1 - e^{-\zeta}\right)^2,$$

$$f_2 = 2\left\{\zeta(1-\zeta)e^{-2\zeta} + e^{-\zeta}\left[\rho^2\zeta^2\left(1 + \rho^2\zeta^2/24\right) - \right.\right.$$

$$\left.\left. - \zeta\left(1 - \zeta/2\right) - \rho^2\zeta^3/2\right]\right\} + (1-\rho^2)\left[\rho^2\left(1 - e^{-\zeta}\right)^2 + \right.$$

$$\left. + 2\zeta e^{-\zeta}\left(1 - \rho^2\zeta/2 - e^{-\zeta}\right)\right].$$

At $\zeta \ll 1$ $f_{1,2} = \zeta^2$ and $\mathcal{F} = \zeta^2(1 - \nu^2 - \nu^4)$, the effectiveness of the conversion η reaches a maximum in the case of the exact resonance ($\nu = 0$). The possibility for a minimum of $\mathcal{F}$ to appear at the exact resonance is associated with the change of a sign of the function f_1. In the case of $\rho < 1.19\,(\varkappa < 0.6)$ at $\zeta = \zeta_1 < 2.4$, the function $f_1(\zeta)$ reverses its sign, and, instead of a maximum of $\mathcal{F}$ at $\nu = 0$, there appears a minimum. Moreover, in the dependence $\eta(\nu)$ instead of one maximum at $\nu = 0$, there appear two ones at symmetric deviations from the TPR. The frequency distance between them is equal to $\sqrt{2(\zeta - \zeta_1) f_1'(\zeta_1)/f_2(\zeta_1)}$ and increases while waves propagate in the region $\zeta > \zeta_1$. It is worth noting that a decrease of $\rho(\varkappa)$ is accompanied by a decrease of ζ_1 and, in particular, at $\rho = 1$ ($\varkappa = 1/\sqrt{3}$) $\zeta_1 = 1.594$ and $f_2(\zeta_1) = \zeta_1^4\left(1 - \zeta_1/2\right)/12$.

With no regard for a variation of the populations of the resonance levels ($n = 1$), expressions (5), (7) still remain valid. That's why the considered decrease of η in the resonance region ($\nu \simeq 0$) is not necessarily conditioned by the saturation effects. As follows from Eq. (1), the minimum in the dependence $\eta(\nu)$ appears as a result of the BRS $\omega_3 \to \omega_1 + \omega_{21}$ related in the optical band to the electron states of atoms or molecules.

At a wide-band pumping, different spectral components of the third harmonic are independent in the considered approximation and, instead of the dependence $a_3(\nu)$, it's possible to consider a spectrum of harmonics while tuning ω_1. In this case, the considered "dip" in the spectrum of the third harmonic is similar to the line of the BRS in the anti-Stokes region with the participation of vibrational states [24]. The possibility of observing the BRS line against the generated radiation is associated with the fact that the effectiveness of parametric processes is determined by the absolute value of nonlinear susceptibility $|\chi_R|$, while the BRS is specified by the imaginary part χ_R'' which decreases more rapidly with increase in ν. At $\varkappa > 0.6$ ($\rho > 1.19$), $f_1(\zeta)$ does not possess negative values, and the BRS line is not formed in the spectrum of the harmonic due to the smallness of the parameter $\varkappa_2$ as compared to $\varkappa_1$.

The obtained conclusions are also proved by the results of a numerical solution of the full system of equations (3). In Fig. 1, the dependences $a_3(\nu)$ and $n(\nu)$ at different values of a_{10} and $\varkappa$ are presented. It is obvious that, for moderate intensities of pumping $a_{10} = 1$, the effects of saturation of the resonance states do not manifest themselves ($n \simeq 1$). Moreover, in the case of $\varkappa \geq 0.58$, there appears a maximum in the resonance region. An increase of the parameter $\varkappa$, i.e. an increase of the ratio $\varkappa_2/\varkappa_1$, is accompanied by the formation of a minimum in the resonance region, which is absolutely stipulated by the BRS. It is interesting to note that, in this case, the difference between the populations of the resonance states n reaches a slight maximum, and the appearance of a minimum of a_3 in the resonance region is not associated with the effect of saturation of the resonance states.

With increasing the pumping intensity ($a_{10} > 1$), the saturation of populations is best observed closely to the exact resonance (see Fig. 1,*b*) and makes an additional contribution to the formation of a "dip" which (unlike the BRS line) can be observed at arbitrary values of $\varkappa$. At $n \ll 1$, the value of χ_R abruptly decreases, which results in a decrease of the effectiveness of the generation of ω_3 in a neighborhood of the resonance. It is worth noting that the minimum in the generation of the harmonic is characterized by a smaller spectral width than that in the difference between the populations n. Thus, a decreasing effectiveness of the resonance generation of the third harmonic under the conditions of the intensive pumping mainly results from the decreasing nonlinearity of the medium due to the equalization of the populations of resonance states [25—27]. The multiphoton ionization that actually decreases the concentration of the nonlinear medium also contributes to the deepening of the minimum in the resonance region [28]. A correct description of its influence requires the account of a change of the PM conditions.

4. Analytic and Numerical Investigation of the Nonlinear Phase Quasi-matching

Let us pass to the consideration of the simultaneous influence of the wave and frequency deviations on the GTH. It is easy to show that the system of equations (3) satisfies the invariance conditions

$$a_{1,3}(s,\nu) = a_{1,3}(-s,-\nu), \quad \theta(s,\nu) = -\theta(-s,-\nu), \qquad (8)$$

which we will consider while analyzing the problem and representing the results.

Under the initial conditions $a_{10} \neq 0$, $a_{30} \simeq 0$ (a_{j0} are the amplitudes of the waves ω_j at $z = 0$), the equation for the generalized phase θ of system (3) contains large terms proportional to a_1^2/a_3 which are stipulated by a change of the phase of the wave ω_3 under the nonlinear parametric interaction. Such members are absent in the amplitude equations. That's why, at the initial stage of the GTH, it is possible to consider rapid variations of θ and slow variations of the wave amplitudes. It is easy to demonstrate that rapid variations of the phase θ in the approximation of fixed amplitudes result in θ possessing the stable value:

$$\theta_0 = \pi/2[1 + \mathrm{sign}(n\varkappa)] + \arctan\nu. \qquad (9)$$

The carried out analytic and numerical investigations demonstrate the stability of the value $\theta = \theta_0$ with respect to small perturbations. Expression (9) can be treated as the initial condition for slow variations of the phase θ and the wave amplitudes $a_{1,3}$. One can show that the value $\theta = \theta_0$ is the optimal one for the process of frequency conversion. In this case, a change of the phase in accordance to (3) is determined by a linear wave mismatch s_L and a nonlinear one $s_{NL} = a_1 n b[3(2\varkappa a_1 + a_3^2/a_1\varkappa) - a_1/\varkappa]/\varkappa$, which is equal to $s_{NL}^0 = n_0 a_{10}^2 \rho b/\varkappa^2$ in the approximation of a specified intensity, the condition $a_3 \ll a_{10}$ being satisfied.

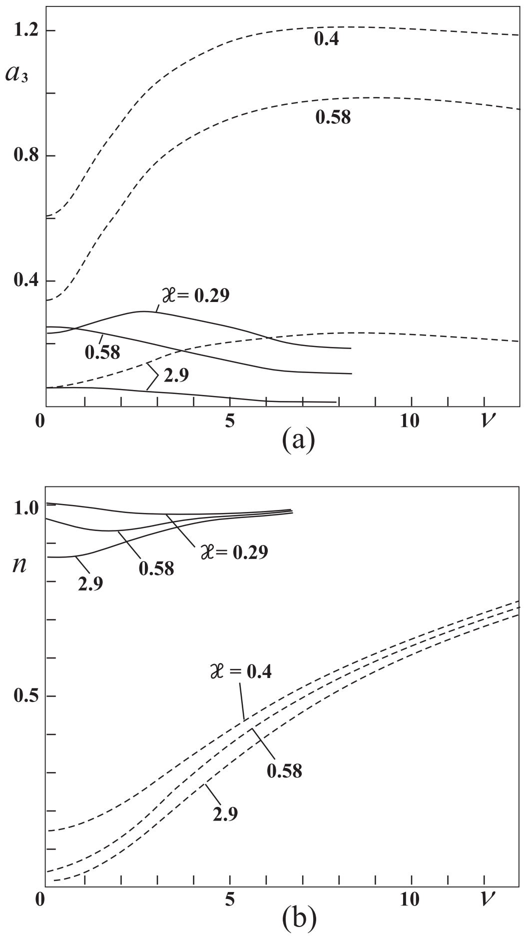

Fig. 1. Dependences of the normalized amplitude of the third harmonic $a_3(\nu)$ (a) and the relative difference of the populations of the resonance states $n(\nu)$ (b) for the dimensionless length of a nonlinear medium $\xi = 0.5$; $a_{10} = 1$ (solid curves), $a_{10} = 3$ (dotted curves) for different values of the parameter $\varkappa$

Under the condition $\zeta \ll 1$, (5) easily gives $\mathcal{F} = a[\zeta^2 - a\zeta^3 + (7a^2 - \beta^2)\zeta^4/12]$, that is the intensity of the third harmonic reaches a maximum at $\beta = 0$, which corresponds to the compensation of the nonlinear wave mismatch s_{NL} by the linear one: $\sigma = -\rho b$. In this case, searching for the extremum of the function $\mathcal{F}(a)$ shows that, at $\zeta < \zeta_0 = 1.256$ $\nu_{\rm opt} = \sigma_{\rm opt} = 0$ and $\mathcal{F}_{\max} = [1 - \exp(-\zeta)]^2$. At $\zeta > \zeta_0$, the maximal effectiveness of the third harmonic is reached at $\nu \neq 0$ and

$$\sigma_{\rm opt} = -\rho\,\frac{\nu}{1+\nu^2}, \quad \zeta_{\rm opt} = \zeta_0\,(1+\nu^2). \qquad (10)$$

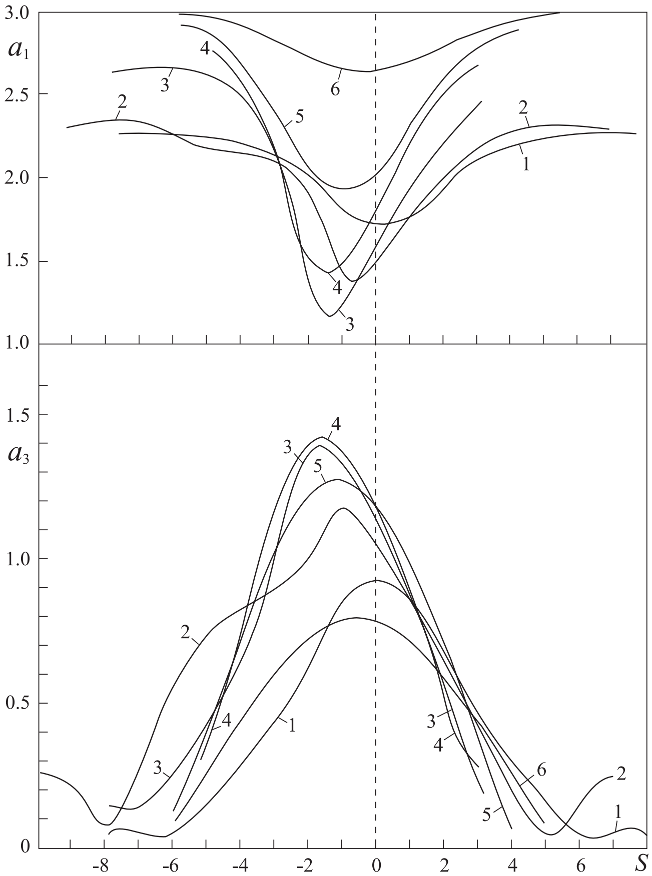

Fig. 2. Dependences of the dimensionless pump amplitudes a_1, a_3 on the normalized wave deviation $s = \Delta n l_0$ in case where $\xi = 1$, $a_{10} = 3$, $\varkappa = 1/\sqrt{3}$ for different values of the frequency deviation from the resonance ν: *1* — $\nu = 0$, *2* — $\nu = 2$, *3* — $\nu = 5$, *4* — $\nu = 10$, *5* — $\nu = 20$, *6* — $\nu = 50$

Moreover, the linear wave mismatch (the term σ in β) can be compensated by a nonlinear wave deviation from the exact TPR. Under the NPM conditions, the frequency ν and wave s deviations for the GTH process have opposite signs at $\rho > 0$ ($\varkappa > 1/\sqrt{6}$) and like signs at $\varkappa < 1/\sqrt{6}$. For $\rho = 0$, the NPM is weakly observed. According to (10), it can be realized only at $|\sigma| \leq \rho/2$ for two possible frequency deviations $\nu_{1,2} = (-\rho \pm \sqrt{\rho^2 - 4\sigma^2})/2\sigma$. The necessary length of the nonlinear medium ζ_{opt} increases in the square law with rising ν due to a decrease of the nonlinear susceptibility χ_R accompanying a deviation from the resonance. For a fixed value of ζ, it is easy to find $|\nu| = \sqrt{\zeta/\zeta_0 - 1}$ and $\eta_{\text{opt}} = \varkappa^2 \left[1 - \exp(-\zeta_0)\right]^2 \zeta/\zeta_0 \simeq 0.407\varkappa^2\zeta$. Thus, under the NPM condition, η does not reach the saturation (like in the case where $s = \nu = 0$), but increases linearly with rising ζ until the used approximation is violated.

It is worth noting the universality of formula (4) and its applicability to different nonlinear processes including those of generating the sum and difference frequencies $\omega_4 = \omega_1 \pm \omega_1' \pm \omega_2$ (ω_1, ω_1' — pump frequencies, ω_2 — the frequency of a signal) under the TPR conditions $\omega_1 \pm \omega_1' = \omega_{21} + \Delta\omega$, as well as the processes of GTH under the conditions of one- ($\omega_1 = \omega_{21}$) and three-photon ($3\omega_1 = \omega_{21}$) resonances [21]. In different cases, it is necessary to change only the expressions for individual quantities, while the general form of expression (4) remains invariable, which proves the universality of the NPM phenomenon.

Since the NPM is substantially related to the phases of interacting waves, it should be considered with regard for the variation of the phase of a pumping wave, i.e. the approximation of a specified intensity should be used. In order to compare, it is worth noting that the approximation of a specified field corresponds to $\rho = -1$, and the signs of the frequency and wave deviations always coincide under the condition of compensation.

The numerical solution of the system of equations (3) with regard for (9) was carried out in a general case of the arbitrarily varying amplitudes and phases of interacting waves. The obtained dependences $a_{1,3}(s)$ in the case of $a_{10} = 3$, $\rho = 1$ ($\varkappa = 1/\sqrt{3}$) at arbitrary values of the frequency mismatch ν are represented in Fig. 2.

Under the conditions of the exact resonance ($\nu = 0$) as in the non-resonance case, the functions $a_{13}(s)$ are even with respect to the wave mismatch s. When introducing the frequency mismatch $\nu > 0$, the maximum of the quantity a_3 and the minimum of the quantity a_1 shift sideways from the negative wave deviations s, as it should be according to (10) for $\rho = 1$. In this case, the dependence $a_3(s)$ can be sufficiently asymmetric, and, along with the central maximum, lateral ones are observed (curve *2*). With increasing a deviation ν from the TPR, the maximum of the amplitude of a generated wave a_3 increases, and the shifting from the conditions of phase matching $s = 0$ continues. For a fixed length of the nonlinear medium $\xi = 1$, the maximal quantum effectiveness of the GTH $\eta_{\max} = (a_{3\max}/a_{10})^2 = 22\%$ is reached at $\nu_{\text{opt}} \simeq 10$ and $s_{\text{opt}} \simeq -1.7$. For $\nu < 0$, according to (8), the maximum of η would be reached at $s > 0$. A further increase of ν is accompanied by both a decrease of the amplitude of the harmonic a_3 and a shift of its maximum in the direction of the PS (curves *5, 6*). Thus, the phase matching represents the boundary case of the NPM.

The pumping intensity decreases due to the GTH and the TAP, that's why the absolute minimum of a_1 is reached at $\nu \sim 5 < \nu_{\text{opt}}$. From Fig. 3,*a* representing the dependence $\theta(s)$ for different ν, it is obvious that, in the case of NPM, the values of $\theta(\xi)$ in the region of maxima of a_3 marked by arrows do not differ essentially from the optimal values $\theta_0(\nu)$ indicated at the axis of ordinates. As a result, the coefficient of power conversion $\eta' = 3\eta$ under the NPM increases from 28% at $s = \nu = 0$ to 66%. In order to compare, it is worth noting that, in the case of $s = \nu = 0$, the maximal conversion coefficient $\eta' = 31.6\%$ can be reached asymptotically with increasing ξ at $\varkappa = 0.675$. It is limited by the parametric wave transparency of matter if $n = 1$ and nonlinear interaction of waves does not occur due to the establishment of the corresponding coherent superposition of waves [17]. Thus, under the NPM, the maximal effectiveness of the GTH per finite length of the medium can be more than twice greater than the asymptotic value of the generation efficiency in the case of the exact resonance.

Previously, the phenomenon of the wave transparency of matter was considered under the conditions of the exact resonance [5—10], but it can also take place in the presence of wave and frequency deviations. Fig. 3,b shows the dependences $n(s)$ for different values of ν. It is obvious that, in the resonance case, the wave transparency determined by the maximum $n \simeq 1$ is observed only in the neighborhood of the PS. The deviation from the resonance $\nu \neq 0$ is accompanied by a shift of the region of the existence of wave transparency from the PS and, in this case, the maxima of n and a_3 (marked by arrows) don't coincide. Fig. 3,b demonstrates a smooth passing from the mode of the wave transparency in the resonance case $s = \nu = 0$ to the non-resonance one where a variation of populations is inessential. It is worth noting that, under the NPM, the region of wave deviations s, where the wave transparency is observed, expands a little and becomes asymmetric.

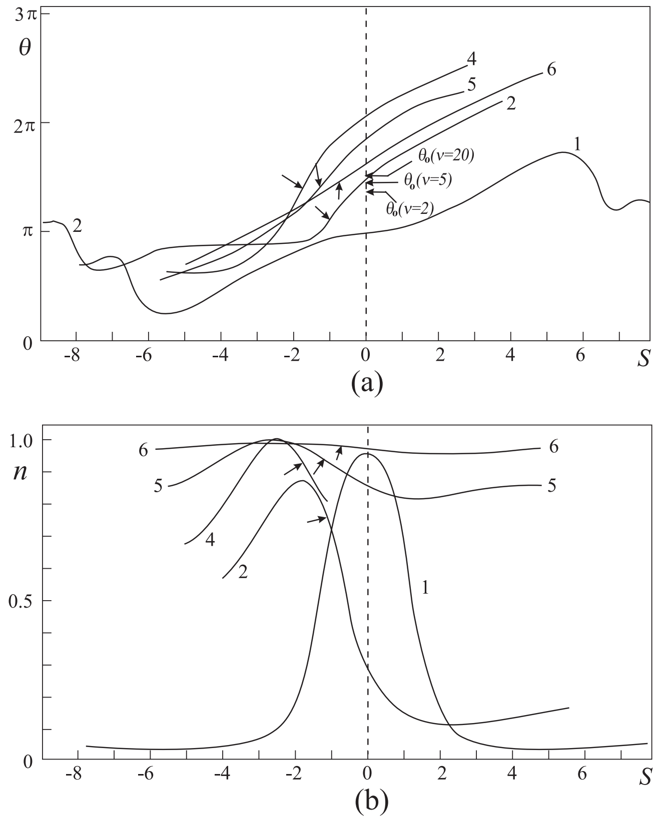

Fig. 3. Dependences of the generalized phase $\theta(s)$ (a) and the normalized difference of the populations of the resonance states n (b) on the normalized wave deviation $s = \Delta n l_0$. Conditions and notations are the same as in Fig. 2. The arrows mark the positions of maxima of a_3 and $\theta_0(\nu)$

Conlusions

The nonlinear quasi-matching is a typical phenomenon of resonance nonlinear optics, which is observed in the third harmonic generation under conditions of the two-photon resonance with respect to pumping as well as in a number of nonlinear parametric interactions of waves and the processes of stimulated scattering (SRS, SBS, etc.). The investigation carried out has demonstrated that the nonlinear wave mismatch qualitatively changes the picture of the nonlinear optical interaction of waves. The maximal efficiency of the generated radiation is reached under the NPM conditions, where the nonlinear wave deviation is compensated by the corresponding linear one. The NPM easily manifests itself in the shifts of the maxima of generated radiation from the exact resonance and phase matching. These deviations clearly demonstrate the variations of the refractive indices induced by strong fields. The well-known phase matching conditions represent the limiting case of the NPM and usually take place in the non-resonance region. The NPM is closely related to the phenomenon of the wave transparency of matter in the presence of the wave and frequency mismatching.

Thus, we have obtained the simple analytic expressions describing NPM, investigated its basic features, and demonstrated their universality. The main rules of the NPM determined analytically are verified by numerical calculations. It is shown that, with a deviation from the exact resonance amounting to 5—10 values of its half-width under the NPM conditions, the effectiveness of the generation of the third harmonic can increase by more than a factor of 2 as compared to the exact two-photon resonance and can reach 66%. In two-photon absorption spectroscopy, a shift of the minimum of the pump radiation sideways from the resonance at the output of the nonlinear medium does not necessarily indicate a shift of the resonance levels but can be conditioned by the simultaneously running process of the GTH.

The results obtained in the present work can be used for explaining the experimental rules observed under resonance parametric interactions and for choosing the optimal parameters of nonlinear media for the third harmonic generation.

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НЕЛІНІЙНИЙ КВАЗІСИНХРОНІЗМ ПРИ ГЕНЕРАЦІЇ ТРЕТЬОЇ ГАРМОНІКИ В УМОВАХ ДВОФОТОННОГО РЕЗОНАНСУ

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Р е з ю м е

Досліджено генерацію третьої гармоніки (ГТГ) в умовах двофотонного резонансу при довільних нелінійних поляризовностях, існуванні хвильової Δk та частотної $\Delta\omega$ відстройки від резонансу з урахуванням зміни заселеностей резонансних станів. Показано, що при нелінійному квазісинхронізмі (НКС), коли нелінійна хвильова відстройка при $\Delta\omega \neq 0$ компенсується лінійною Δk, ефективність ГТГ може зростати приблизно у 2 рази (порівняно з випадком $\Delta k = \Delta\omega = 0$) і перевищувати 60%. Встановлено, що наближення заданого поля накачки не дозволяє описати явище НКС і необхідно користуватися наближенням заданої інтенсивності. Аналітично та чисельно показано, що при НКС хвильова і частотна відстройки можуть мати як однакові, так і різні знаки. У спектрі гармоніки може з'явитися "провал", зумовлений оберненим комбінаційним розсіянням, а насичення заселеностей резонансних станів спричинює його поглиблення при великих інтенсивностях накачки.