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Possibility of creating optical parametric oscillators with a precise shift of oscillation frequency, excited by monochromatic signal injection

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Abstract. **The approximation of interacting quasi-monochromatic plane waves is used to analyse the possibility of parametric generation of pulses with a frequency shift relative to the injected monochromatic signal. This problem is important for the development of receiver–transmitters with heterodyne detection for remote IR spectroscopy of the atmosphere with a high spectral resolution and a long-range probing. Such systems can be fabricated on the basis of recently developed pulsed and cw optical parametric oscillators. The main attention is paid to the hitherto uninvestigated problem of nonadiabatic excitation of oscillations with a shift in the injected signal frequency relative to the cold resonator mode frequencies. It is shown that the oscillation frequency (frequencies) can be controlled upon such an excitation without increasing the spectral linewidth. The conditions for efficient excitation of single-frequency and two-frequency oscillations at adjacent resonator modes are found. Conditions for sufficient frequency stabilisation are determined.**

Keywords: *optical parametric oscillations, remote spectroscopy, signal injection.*

1. Introduction

Optical parametric oscillators (OPOs) excited by signal injection (SIOPOs) emit pulses with a small spectral width (see, for example, Refs [1–12]). Upon excitation by a monochromatic signal, these pulses can be transform-limited (see, for example, Refs [7–9]). A precise tuning of the oscillation frequency of SIOPOs relative to the frequency of the monochromatic light being injected broadens the scope of their application in linear and nonlinear spectroscopy. However, the possibility of controlling the frequency shift of such oscillators has not been studied so far upon nonadiabatic pumping, when tuning may occur without a broadening of their emission spectrum.

A decrease in the width of the emission spectrum of OPOs pumped by a narrowband radiation was demonstrated in 1969 in Ref. [1]. A theoretical analysis of the problems related to such an excitation of OPOs was carried out in Refs [2–7] in the approximation of interacting quasi-harmonic plane waves. In Refs [8–12], the excitation of oscillations by injection of a monochromatic signal was simulated numerically by taking into account the relative drift and diffraction of interacting beams, depletion of the pump wave, as well as the group detuning of interacting waves.

Several experimental works (see, for example, Refs [6–12] and references therein) have confirmed the theoretical conclusions concerning the conditions of narrowing of the parametric oscillation spectrum and an increase in the conversion coefficient due to the signal injection. For example, transform-limited nanosecond pulses with the corresponding spectral width of the order of 10^{-2} cm^{-1} were obtained in Refs [7–9].

In the above-mentioned studies, a deviation of the injected signal frequency ω_s from the natural frequency ω_{10} of the nearest longitudinal mode of the 'cold' OPO resonator did not exceed the spectral width of the mode and one percent of the mode interval Δ_m. A noticeable shift of the oscillation frequency ω_1 relative to ω_s occurs, for example, for a finite wave detuning due to the dependence of ω_1 on the pump intensity and the intensity of the exciting waves. For real values of the parameters, the shift does not exceed $0.1\Delta_m$. Such an excitation of SIOPO can be called quasi-adiabatic because the pumping efficiency in such a case depends weakly on the steepness of the leading edge of the pump pulse. It was mentioned in Refs [7–9] that a shift of the lasing frequency due to quasi-adiabatic mechanisms inevitably leads to a certain broadening of the spectrum caused by the phase modulation of the emission intensity associated with the dependence of the wave intensity on time and transverse coordinates. Moreover, instability of the peak pump power will lead to an instability of the OPO frequency.

The above-mentioned effects, which virtually rule out a precise tuning through quasi-adiabatic processes, can be avoided if lasing is excited upon zero detuning from synchronism by injecting a signal whose frequency has the required shift relative to the frequency of the nearest mode of the 'cold' OPO resonator. For a sufficiently steep leading edge of the pump pulse, the pump efficiency will depend weakly on the pump frequency shift $\Omega_s = \omega_s - \omega_{10}$. In this case, lasing will occur virtually at a single frequency $\omega_1 = \omega_{10}$ as long as the frequency shift $\Omega_s < 0.45\Delta_m$. If, however, the frequency of the signal being injected is somewhere in the middle between frequencies of the adjacent modes of the OPO resonator ($|\Omega_s - 0.5\Delta_m| \leqslant 0.02\Delta_m$), the main part of the output (at least 75 %) will be concentrated in these

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adjacent modes. Such a two-frequency lasing mode is interesting, for example, for remote spectroscopy of differential absorption using a single receiver–transmitter.

The importance of the task of fabricating OPOs pumped by injecting a signal with a shift of the oscillation frequency (or frequencies) without broadening the emission spectrum is associated to a considerable extent with the possibility of their application for creating receiver–transmitters with heterodyne detection (RTHD) for remote spectroscopy of the atmosphere in the near- and mid-IR ranges. This problem has been solved owing to the advances made in recent years in the fabrication technology of nonlinear elements with a periodic polarisation and to the achievements based on the use of these elements for fabricating cw pumped OPOs tunable in the near- and mid-IR ranges [13–18]. These OPOs can serve as RTHD heterodynes and as pump sources for SIOPOs.

The sensitivity of heterodyne receivers may be close to the limiting value (see, for example, Ref. [19]), i.e., it may nearly achieve a photon in a time equal to the duration of a probing pulse. In the mid-IR range for a pulse duration of ~ 10 ns, this is about 2–3 orders of magnitude higher than the sensitivity of direct detection receivers. The use of lidars with RTHD in this spectral range will considerably broaden the potentialities of remote spectroscopy of the atmosphere. For example, the conversion coefficient of lithium niobate OPO may be as high as tens percent, and the probing pulse energy may be tens of millijoules for an appropriate choice of the optical scheme and its parameters [20, 21].

Thus, in spite of a decrease in the scattering coefficient $\sim 1/\lambda^4$, lidars with RTHD can detect, for example, radiation scattered by high atmospheric layers (up to 20–40 km), where the scattering is weakened due to a decrease in the concentration of aerosols. This makes it possible to monitor at these heights a number of molecular components (e.g., CH_4, NH_3, HCl, NO_2), which play an important role in the physicochemical processes occurring in the atmosphere, from earth or through satellites. At present, such a monitoring is possible only with the help of aircrafts. We studied excitation of SIOPO in this paper for parameters that were close to optimal values for a $LiNbO_3$ SIOPO pumped by Nd:YAG laser pulses.

2. Equations for oscillations in a single-resonator travelling-wave OPO

The energy characteristics of parametric oscillations depend significantly on the type of OPO resonator, transverse size of the pump beam, and the angles between the group velocities of the interacting waves. If oscillations are excited by signal injection, the spectral characteristics of oscillations and the pump efficiency willl also depend on these factors. However, their characteristic dependences on detuning of the injected signal frequency relative to the natural frequencies of the resonator, as well as detuning from synchronism and steepness of the leading edge of the pump pulse can be determined by considering the simple model of the OPO in the approximation of interaction of plane quasi-monochromatic waves.

Consider in this approximation the excitation of a single-resonator OPO by injecting a monochromatic signal whose frequency ω_s lies between the natural frequencies ω_{10} and ω_{11} of two adjacent resonator modes. The fields $\boldsymbol{E}_1$, $\boldsymbol{E}_2$ of resonance and nonresonance waves, and $\boldsymbol{E}_3$ of the pump wave in a nonlinear crystal can be written in the form

$$\boldsymbol{E}_i = \boldsymbol{e}_i[A_i(z,t)\exp \mathrm{i}(k_i z - \omega_{i0} t) + \text{c.c.}]. \tag{1}$$

Here, ω_{30} is the pump frequency; $\omega_{20} = \omega_{30} - \omega_{10}$ is the frequency of the nonresonance wave; k_i are the projections of the wave vectors on the resonator axis z, corresponding to the frequencies ω_{i0}. In this notation, the amplitude of the injected signal incident on the input mirror of the resonator is described by the expression

$$A_s(t) = m(\cos \Omega_s t - \mathrm{i} \sin \Omega_s t), \tag{2}$$

where $\Omega_s = \omega_s - \omega_{10}$.

The amplitudes A_i and A_s are assumed to be normalised in such a way that the photon flux density of the corresponding waves is equal to the square of their modulus multiplied by the maximum pumping photon flux density n_p during the pulse, which is proportional to the peak intensity $I_p = n_p \hbar \omega_{30}$. We will confine our analysis to the cases in which the spectral width of the excited oscillations is much smaller than the spectral width Δ_{12} of the phase-matching for the resonance wave. In the equations for the amplitudes of waves interacting in a nonlinear crystal under these conditions, their group velocities can be assumed to be equal to the velocity v of the resonance wave. In the given quasi-static approximation, the interaction between the waves will be determined by two parameters: the wave detuning $\Delta k = k_1 + k_2 - k_3$ from synchronism, and the interaction coefficient γ. For the chosen normalisation of amplitudes, we have $\gamma = \chi\sqrt{I_p}$, where χ is the nonlinearity parameter of the crystal.

In the quasi-static approximation, the solution of the problem on wave propagation in a nonlinear crystal is reduced to the solution of known (see, for example, Ref. [22]) nonlinear equations in ordinary derivatives for amplitudes $A_i(z,\tau)$, where $\tau = t - z/v$. For identical attenuation of waves, these equations have analytic solutions, which can be expressed in terms of Jacobi functions [22]. However, it is easier to calculate the dependence of amplitudes $A_i(d,t)$ at the output of a crystal of length d on the amplitudes $A_i(t)$ at its input surface by solving the equations numerically. For a more visual presentation of the parameters determining these dependences, we write the equations for $A_i(z,t)$ in an integral form, neglecting for simplicity the decay of waves, which does not affect the results of computations:

$$A_{1,2}(z,t) = A_{1,2}(\tau) + \mathrm{i}\Gamma \int_0^\zeta \mathrm{d}\xi A_3(\xi d,\tau) A_{2,1}^*(\xi d,\tau) \exp(-\mathrm{i}\xi\Delta),$$

$$A_3(z,t) = A_3(\tau) + \mathrm{i}\Gamma \int_0^\zeta \mathrm{d}\xi A_1(\xi d,\tau) A_2(\xi d,\tau) \exp(\mathrm{i}\xi\Delta). \tag{3}$$

Here, $\zeta = z/d$; $\Gamma = \gamma d$ is the amplification parameter; $\Delta = \Delta k d$ is the detuning from synchronism.

In a single-resonator OPO, we have $A_2(t) = 0$ at the input surface of a nonlinear crystal, and, hence, it follows from (3) that the resonance wave amplitude at the output surface of the crystal is determined by the quantities $A_1(t)$ and $|A_3(t)|$ at the instant of time $t - d/v$ with the help of the successor function $U_1[A_1; |A_3|]$. This function can be determined either analytically (see, for example, Ref. [22]) or numerically directly from Eqns (3):

$$A_1(d,t) = U_1[A_1(t-d/v); |A_3(t-d/v)|]. \quad (4)$$

In the case under study, the phase modulation of the pump intensity is transformed into phase modulation of a nonresonance wave and does not affect pumping. This allows us to assume that $A_3(t)$ is a real quantity.

We shall confine our analysis to the case when the OPO resonator does not contain any dispersion element. Without loss of generality, we can assume that the input mirror of the resonator coincides with the input surface of the crystal. In this case, taking into account the choice of the reference system for the frequency shift, the equation for the amplitude $A_1(t)$ can be written in the form

$$A_1(t) = (1-R_0)^{1/2} A_s(t) + \sqrt{R} U_1[A_1(t_L); A_3(t_L)]. \quad (5)$$

Here R_0 is the reflection coefficient of the input mirror; $R = R_0 K$; $K \leqslant 1$ is the transmission coefficient in the resonator; $t_L = t - T$; $T = L/v$ is the transit time for a resonance wave in the resonator of optical length L.

The most promising resonators for use in RTHD are travelling-wave resonators (in particular, three-mirror resonators), in which the output mirror coincides with the input mirror, and $K \approx 1$. We shall consider this case below, assuming that $K = 0.95$.

3. Analysis of excitation

Some general conclusions about the excitation of parametric oscillations may be drawn from an analysis of the dependence of the resonance wave amplitude at the output of the crystal on its amplitude A_1 at the input surface for a constant pump amplitude $A_3(t) = 1$. Fig. 1 shows the dependences of the amplitude transfer function $V_1 = |U_1|$ and of the sine of the phase incursion $\Delta\varphi_1$ of a resonance wave upon its amplification (phase of the function U_1) on the modulus of A_1 for several characteristic values of parameters Γ and Δ.

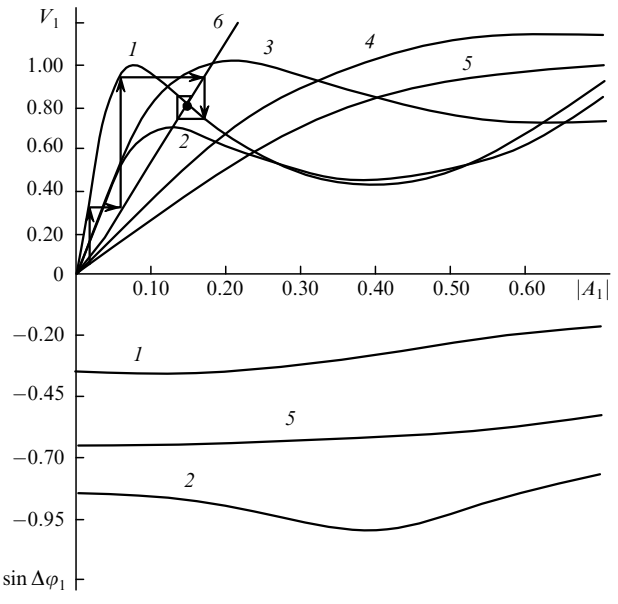

Figure 1. Dependence of the transfer function V_1 and $\sin\Delta\varphi_1$ on $|A_1|$ for $\Gamma = 4$ (*1*, *2*), 3 (*3*–*5*), and $\Delta = 0$ (*3*, *4*), 1 (*1*), 3 (*5*), 6 (*2*), as well as the straight line $V_1 = |A_1|\sqrt{R}$ (*6*).

The points of intersection of the transfer function with the straight line $V_1 = |A_1|/R^{1/2}$ determine, as is known, the amplitude of steady-state oscillations for the corresponding values of R. The broken line with arrows tending to these points from the initial point $V_1, A_1 \approx 0$ describes the establishment of the oscillation amplitude when pump is instantaneously switched on. The frequency observed upon excitation of oscillations is related to the dependence of the phase incursion $\Delta\varphi_1$ on the oscillation amplitude for a finite wave detuning. This dependence determines the variation of the instantaneous frequency of the excited oscillations: $\Delta\omega = \Delta\varphi_1/T$.

One can see from Fig. 1 that $\Delta\varphi_1$ depends weakly on the oscillation amplitude for $|A_1| < 1$, $\Gamma \geqslant 2$ and $\Delta \leqslant 6$. This dependence may become significant only for values of the amplification parameter Γ and the attenuation coefficient R close to unity. Hence, upon pulsed pumping in the cases of interest, $\Delta\varphi_1$ can be assumed to depend only on detuning Δ and pump intensity, i.e., $\Delta\omega$ can be determined from the expression obtained in the linear approximation in the amplitude of the generated waves:

$$\Delta\omega = \frac{\Delta\varphi_1}{T}, \quad \Delta\varphi_1 \approx -\frac{\Delta}{2}\left(1 - \frac{\tanh G}{G}\right),$$
$$G = \Gamma A_3 \left[1 - \frac{\Delta^2}{4(\Gamma A_3)^2}\right]^{1/2}. \quad (6)$$

It follows from these equations that the maximum value of the frequency shift of the oscillations will be observed in the region of the highest pump intensity, and their spectrum will be broadened nonsymmetrically because of a decrease in the value of $\Delta\varphi_1$ at the pump pulse edges.

To estimate the spectral broadening and to find the dependences in which we are interested, Eqns (3)–(5) were solved numerically. The pump pulse shape was written in the form:

$$A_3(t) = \tanh^2\frac{t}{T_f}\cos^2\frac{\pi t}{4T_p} \text{ for } 0 < t < 2T_p,$$

$$A_3(t) = 0 \text{ for } t > 2T_p.$$

For $0 \leqslant t \leqslant T$, the amplitude $A_1(t)$ was assumed to be equal to the amplitude of the steady-state oscillations in the cold resonator:

$$A_1(t) = m\frac{(1-R_0)^{1/2}[(1-\sqrt{R}\cos\varphi) + i\sqrt{R}\sin\varphi]}{1 - 2\sqrt{R}\cos\varphi + R}$$
$$\times \exp(-i\Omega_s t). \quad (7)$$

Here, T_p is the pump pulse duration; T_f is the duration of the leading edge of the pump pulse; $\varphi = 2\pi f_s$; $f_s = \Omega_s T/2\pi$ is the detuning of the injected signal frequency, measured in units of the mode interval. We obtained the dependences of the instantaneous conversion coefficient $I_1(t) = R_0|A_1(d,t)|^2$, and the quantity

$$J_h(\eta) = \{\cos[2\pi(f_s + f_0)\eta]\mathrm{Re}A_1(d,\eta) - \sin[2\pi(f_s + f_0)\eta]\mathrm{Im}A_1(d,\eta)\}.$$

on the dimensionless time $\eta = t/T$.

We will call the quantity J_h the intermediate-frequency signal because it is proportional to the variable component of the current of a photodetector exposed to a part of the emitted radiation and to the monochromatic signal from the heterodyne with a frequency shift $\Omega_0 = 2\pi f_0/T$ relative to the injected signal. The spectrum of J_h is related to the emission spectrum in an obvious way. We will calculate the modulus of the spectrum $J_h(\eta)$ and will call it the signal spectrum $J_h(f)$. Note that the spectral intensity is proportional to the square of this quantity.

Fig. 2 shows the dependences $I_1(\eta)$ and $J_h(\eta)$ illustrating the frequency shift and the emergence of phase modulation of the emitted radiation for a finite wave detuning. The spectra of the amplitude $J_h(\eta)$, calculated for some values of the parameters Δ, Γ and f_s are shown in Fig. 3. One can see from the dependences that the shift in the oscillation frequency due to a finite wave detuning leads to a non-symmetric spectral broadening of the order of a few tenths of percent of the frequency shift $\Delta\omega$.

For a zero wave detuning Δ and a finite detuning of Ω_s, no spectral broadening is observed (see Fig. 3). If the detuning of Ω_s is sufficiently small compared to the mode interval ($|f_s| \leqslant 0.4$), oscillations are excited at the frequency ω_{10} of the nearest mode. Random variations in the pump frequency or other factors like crystal temperature variations lead to slight changes in the detuning Δ and, in accordance with (6), to small shifts $\Delta\omega$ of the oscillation frequency relative to ω_{10}. Relations (6) make it possible to estimate the required stability of the pump frequency and other parameters leading to random variations in $\Delta\omega$.

An increase in the frequency detuning f_s lowers the excitation efficiency of oscillations. This decrease and its rate depend to a considerable extent on the duration T_f of the leading edge of the pump pulse. This dependence was analysed at $T_p \to \infty$. First, the time t_{n0} of the onset of nonlinear regime was determined for $f_s = 0$ and for given values of T_f and the amplitude $m = 10^{-6} - 10^{-9}$ of the injected signal. Then, the amplitude $m(f_s)$ of the signal for $f_s > 0$ was selected in such a way that the time $t_n(f_s)$ of the onset of nonlinear regime differs from t_{n0} by no more than $T/2$. The dependence of the pump efficiency in amplitude $M(f_s) = m/m(f_s)$ is shown in Fig. 4 for several values of T_f. These dependences vary insignificantly upon a variation of the amplification parameter Γ from 2 to 4 and the loss coefficient R from 0.5 to 0.7. The dependences shown in Fig. 4 correspond to $\Gamma = 3$ and $R = 0.5$. For some values of the parameters, only the extreme points for $f_s = 0.5$ are shown.

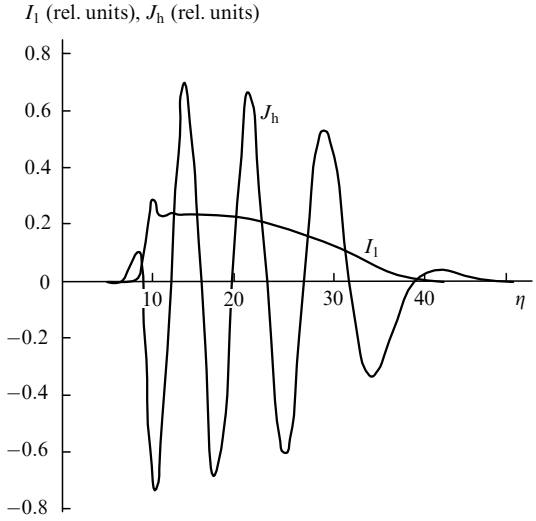

Figure 2. Dependences $I_1(\eta)$ and $J_h(\eta)$ for $T_f/T = 4$, $T_p/T = 25$, $\Delta = 4$, $f_s = 0$, $m = 10^{-4}$.

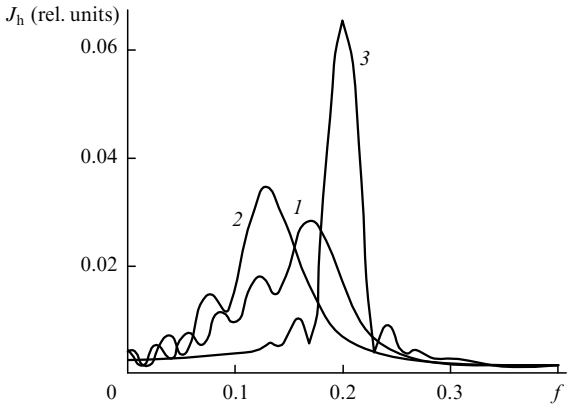

Figure 3. Spectra $J_h(f)$ for f/T = 4, $T_p/T = 25$, $f_0 = 0$, $m = 10^{-4}$ and $f_s = 0$ (*1*, *2*) and 0.2 (*3*), $\Delta = 4$ (*1*, *2*) and 0 (*3*), $\Gamma = 4$ (*1*) and 3 (*2*, *3*).

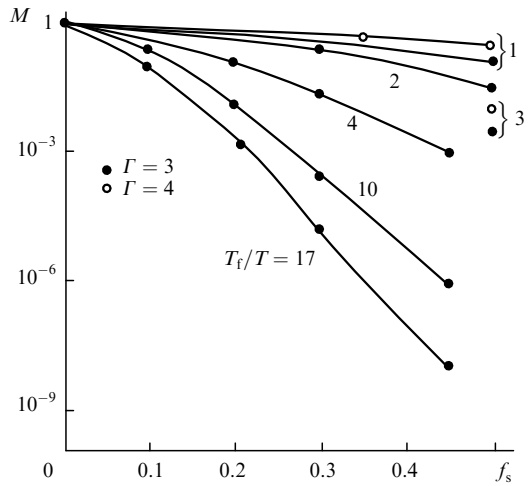

Figure 4. Dependence of the pump efficiency M on f_s for different values of T_f/T and Γ.

It follows from the data presented in Fig. 4 that for values of the nonadiabaticity parameter $N_a = |f_s|T_f/T$ not exceeding unity, the pump efficiency M changes weakly. In this case, the value of M decreases due to an increase in the parameter N_a and due to the dependence of the amplitude of the steady-state oscillations (7), excited before the arrival of the pump pulse, on f_s. Upon a further increase in the nonadiabaticity parameter, the pump efficiency decreases rapidly due to a weakening of the transformation of induced oscillations into natural oscillations of the oscillator.

As the frequency of the injected signal approaches the middle of the mode interval ω_{10}, ω_{11}, oscillations are excited

not only at the frequency closest to ω_s, but also at the frequency ω_{11} of the adjacent mode. At the nonlinear stage of the development of oscillation, the oscillations of the intensity [which is proportional to $I_1(t)$] decay over several transits through the resonator in the same way as shown in Fig. 2, while the oscillations of $J_h(t)$ become considerably nonsinusoidal, i.e., quasi-periodic with a period close to $2T$ (for $f_0 \ll 1$). This means that after excitation of two adjacent modes, their interaction at the nonlinear stage leads to excitation of other modes (see Fig. 5b), so that the phase of the emitted radiation will be modulated with the intermode frequency. It follows from the above calculations that the main part of the energy (at least 75 %) will be concentrated at frequencies ω_{10} and ω_{11}.

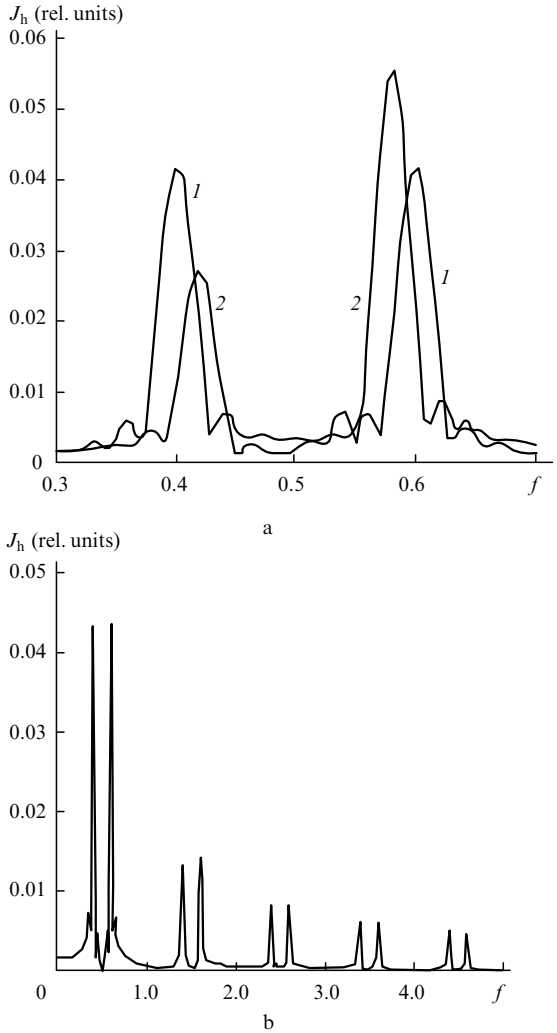

Figure 5. Spectra $J_h(f)$ for $\Gamma = 3$, $T_f/T = 2$, $T_p/T = 25$, $\Delta = 0$, $f_0 = 0.1$, $m = 10^{-4}$, $f = 0.5$ (*1*), 0.48 (*2*) and 0.5 (b).

Such a nearly two-frequency lasing can be used for differential absorption spectroscopy if the relative intensity at frequencies ω_{10} and ω_{11} does not change significantly in the course of measurements, and the intermediate frequencies fluctuate within no more than the linewidth. In this case, the heterodyne frequency must differ from the frequency ω_s by Ω_0, exceeding the linewidth. It follows from the data presented in Fig. 5 and Table 1 that these requirements are met for an instability of the resonator frequencies ω_{10} and ω_{11} relative to ω_s not exceeding 1 % – 2 % of the mode interval. The data presented in Table 1 and Fig. 4 also show that a high efficiency of excitation of two-frequency lasing and its high stability can be ensured only with the help of pump pulses with a steep leading edge with duration $T_f \simeq (1-4)T$. Such pulses can be easily formed with the help of Pockels cells or in multipass amplifiers with an SBS mirror (see, for example, Ref. [23]).

Table 1. Dependence of the relative intensity of adjacent modes $G(A_{11}/A_{10})^2$ on the parameters T_f/T, f and Γ.

f	Γ	$T_f/T = 1$	$T_f/T = 2$	$T_f/T = 4$
0.495	4	0.73	0.6	0.41
	3	0.7	0.56	0.26
0.49	4	0.58	0.4	0.21
	3	0.53	0.39	0.17
0.48	4	0.42	0.25	0.08
	3	0.36	0.21	$< 10^{-2}$
0.45	4	0.19	0.06	$< 10^{-2}$
0.4	4	0.06	$< 10^{-2}$	

4. Conclusions

The above analysis shows that transform-limited pulses with frequency (or frequencies) shifted relative to the frequency of the injected signal can be produced by using nonadiabatic excitation of single-resonator OPOs with a pulsed pumping through a monochromatic signal injection. The closeness of the injected signal frequency and the pump radiation frequency to the corresponding frequencies of exact phase matching serves as a condition for attaining an extremely small width of lasing lines and a stability of their frequency. For example, this condition will be satisfied in the case of excitation of 20 – 40-ns pulses (corresponding to a limiting spectral width of the order of 10^{-3} cm^{-1}) if the spectral width and instability of the pump laser frequency do not exceed 10^{-1} cm^{-1}. Such values of the parameters are achieved in modern lasers.

The analysis was carried out in the approximation of interacting plane waves. In real systems with finite transverse dimensions of the interacting beams, the above conclusions will be valid only for interactions of the fundamental longitudinal modes of the OPO resonator because the wave intensity variations are not transformed into phase variations of the oscillations in the case of exact phase matching. However, for an inadequate matching of the transverse structure of the injected signal with the OPO resonance mode structure, higher transverse modes will be also excited due to the nonadiabaticity of the process, which may lead to a broadening of the emission spectrum and to a lowering of the stability of its frequency. This effect can be avoided by using the OPO with a high degree of suppression of the excitation of higher transverse modes. The development of such OPOs with a high pulse energy and a high efficiency is an independent problem, which may be solved as a result of subsequent investigations.

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