THE CONCEPT OF MULTIPLE NONCRITICAL PHASE-MATCHINGS ON A NON-LINEAR FREQUENCY CONVERSION IN BIAXIAL CRYSTALS

N.E. KORNIYENKO, V.I. ZADOROZHNII, T.S. SIDENKO

Taras Shevchenko Kyiv National University
(2, Academician Glushkov, Kyiv 03680, Ukraine; e-mail: nikkorn@univ.kiev.ua)

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A general approach to the analysis of conditions of the noncritical phase matchings (PMs) of various multiplicities in the process of generation of a summary frequency is developed. As an example, the analysis is applied to a KTP (potassium titanyl-phosphate) biaxial crystal. The approach is based on both the consideration of general properties of the PM surface constructed in the coordinates of the frequencies and the propagation angles of interacting waves and the analysis of their singular lines and the intersection points of these lines. On the PM surface, we consider the lines of noncriticality in the signal frequency or the propagation angles of the pumping wave and the signal one. A classification of twice noncritical PMs (in the frequency and one angle or in two angles) is proposed. It is found that the region of double noncriticality in the frequency and the IR signal divergence can be scanned over the whole transparency region of the KTP crystal on the tuning of the frequency of a long-wavelength laser pumping. A new type of the multiple noncritical synchronism (conditional PM) that is realized on the consistent change in the directions of the pumping and signal waves in the region of the maximum angle between the interacting waves is analyzed. Is is shown that the noncritical matching can be realized in three independent parameters — frequency and two angles. The use of the multiple noncritical PM allows the visualization of wide-band IR spectra and IR images and the frequency conversion of femto-second laser pulses.

1. Introduction

The concept of phase matching (PM) is important in the physics of the non-linear wave interaction and in modern optics. When the conditions for the PM are fulfilled, one can observe the effective frequency conversion even at small non-linearities in the crystal transparency bands. The angular and frequency widths of the PM are determined by linear terms in the expansion of the wave mismatch $\Delta\mathbf{k}$ in the frequency and angles. In the case of such critical PM, the widths of the frequency ($\delta\,\nu$) and angular ($\delta\varphi$) distributions of the PM are quite small (several cm $^{-1}$ and tens of angular minutes). Due to this fact, the resulting radiation contains the information encoded in the frequency-angular distribution (and hence in the spatial and temporal shapes) of the signal radiation. This is used for the visualization of the IR and UV signals and images, since the high-speed, wide-band, and multichannel radiation detectors, including highly sensitive CCD cameras, are available in the visible and near IR ranges. Just the feasibilities of practical applications of the signal conversion on the basis of the generation of the summary and difference frequencies $\omega_R = \omega_P \pm \omega_S$ (the subscripts P, S, and R refer to the pumping, signal, and generated waves, respectively) attract the long-lasting attention to the investigation of these processes [1—18].

For the purposes of non-linear spectroscopy, it is desirable to have the signal conversion in a range of frequencies and angles to be as wide as possible [3,5]. The problem of the formation of wide PM bands is important also in the frequency conversion of the pico- and femto-second laser pulses by the methods of non-linear optics and in the study of fast processes [10—12]. For example, one needs a PM frequency bandwidth $\delta\,\nu \sim 200$ cm $^{-1}$ in order to work with pulses of $\sim$ 50-fs duration and the bandwidth of 1300 cm^{-1} [9] for the shortest experimentally observed $\sim$ 8-fs pulses. Under these requirements, we encounter a task of creating the special (noncritical) conditions for the PM, at which the linear terms of the expansion of the wave mismatch $\Delta\mathbf{k} = \mathbf{k}_R - (\mathbf{k}_P \pm \mathbf{k}_S)$ in a series in the frequencies and angles vanish, and the quantities $\delta\nu$ and $\delta\varphi$ are determined by the quadratic terms of this expansion.

In order to increase the angular aperture of a non-linear frequency conversion in anisotropic crystals and in isotropic liquid and gaseous media, the popular vector scheme of the tangential synchronism (the Warner scheme) [1, 2] or the collinear wave interaction scheme at 90° PM is usually used. The wide-band frequency conversion is realized with the use of the group synchronism [1,4,7, 13—17,23,25], when, along the usual conditions of the vector PM, the condition of matching the group speeds of the signal and resulting radiations is imposed. Under a monochromatic pumping, this can provide the fulfillment of the ordinary conditions for the PM in a wide frequency range. In particular, in the collinear case, the condition of the equality of the group velocities, $\partial\omega/\partial k_S = \partial\omega/\partial k_R$, for the central frequencies of the signal and the generated radiations should be fulfilled. For the vector group synchronism, the problem is reduced to matching the anisotropy and the group velocities of interacting waves, which is discussed later in this paper.

Particular cases of the nonlinear wave interaction, being noncritical in frequencies and angles, were implemented in works [1, 3, 4, 7,23—25]. The frequency range of the group synchronism can be tuned by changing the pumping wavelength or by the use of various non-linear crystals and vector wave interactions [4, 7]. Especially we should notice the simultaneous implementation of the group and tangential PM, i.e. the double noncritical PM in the frequency and angle [1, 5]. In this case, for the pumping at $\lambda_P = 1.064$ μm, the values $\delta\nu = 260$ cm^{-1} and $\delta\varphi = 35$ mrad were obtained in a $LiNbO_3$ crystal near $\lambda_S = 3.5$ μm. The combination of the two adjacent group PMs in the range 9–14 μm with the use of two non-linear crystals $AgGaS_2$ and $HgGa_2S_4$ and one pumping wavelength was considered in [13], but the problem of vector group and multiple noncritical PMs (in two or three parameters) remained for a long time insufficiently studied in the general aspect.

We have proposed a general concept of the realization and classification of the multiple (double and triple) noncritical PMs in frequencies and angles [14—16]. This approach is based on the analysis of the general properties of a PM surface constructed in the coordinates of frequencies and angles and on the analysis of its singular lines and points. For the first time, the feasibility of scanning the double noncriticality region (in frequencies and angles simultaneously) is demonstrated for the frequency conversion in the whole transparency band of non-linear crystals. We have discovered an interesting physical phenomenon of the triple noncritical PM (in the frequency and two angles of the propagation of the pumping and signal waves). The region of the triple noncritical PM corresponds to the end point of the line of double noncriticality. The neighborhood of this point resembles the critical point in a liquid-gas diagram. This is why the considered problems are of interest not only in the fields of quantum electronics and non-linear optics, but can be used to describe the critical states of matter from the qualitatively new viewpoint. The phenomena related to noncritical PM are important in the energy relaxation in condensed matter and in the analysis of the spectra of the second and higher orders in vibrational spectroscopy. In our previous works [14,15], a more attention was paid to the demonstration of the potentialities of noncritical PMs in uniaxial crystals. The goal of this work is the investigation of the new types of multiple noncritical PMs, the frequency scanning of the region of double noncriticality, and the determination of the frequencies of triple noncritical PMs in various crystallographic planes of a biaxial KTP crystal.

2. Analytical Consideration of PMs Noncritical in the Frequency or Angles

In a non-linear crystal, the parametric interaction of waves is most efficient under the PM conditions

$$\omega_R = \omega_P \pm \omega_S,$$

$$\Delta\,\mathbf{k} = \mathbf{k}_P\,(\omega_P, \theta_P,\,\Phi_P) \pm \mathbf{k}_S\,(\omega_S, \theta_S,\,\Phi_S) -$$

$$-\mathbf{k}_R\,(\omega_R, \theta_R,\,\Phi_R) = 0, \qquad (1)$$

where the angles θ_j, Φ_j ($j = P, S, R$) determine the wave propagation directions relative to the principal optical X, Y, Z axes of the crystal (Fig. 1). The KTP crystal belongs to the orthorhombic symmetry class $mm2$. Its optical axes lie in the ZX plane, with the Z-axis being a bisector of the angle between the optical axes. The main refractive indices $N_{X,Y,Z}$ satisfy the inequality $N_Z > N_Y > N_X$. Equations (1), which take into account the dispersion law $\mathbf{k}_j = \mathbf{k}_j(\omega_j, \theta_j, \Phi_j)$, describe the PM surface in coordinates ω_j, θ_j, and Φ_j. The topology of this surface determines the angular and frequency structure of a generated radiation [14—16]. The shape of the PM surface corresponding to the propagation of three waves in an arbitrary plane of the biaxial crystal is quite complicated. However, the general characteristics, which are important for applications, of the frequency-angle structure change on the parametric frequency conversion can be understood from the consideration of the characteristic features of the PM surfaces for vector wave interactions in the main crystal planes XZ, YZ, and XY, as is considered below.

Consider the monochromatic pumping ($\Delta\omega_P = 0$), which unambiguously determines the frequency ω_R at the given values ω_P and ω_S. In this case, for each fixed pumping frequency ω_P, the vector equation of a PM surface under the wave interaction in a given plane can be replaced by the equivalent system of two independent scalar equations. These are the equations for the longitudinal and transverse components of $\Delta\,\mathbf{k}$ for a given direction (for example, $\mathbf{k}_R$) that relate the signal wave frequency and the angles of wave propagation in the selected main plane:

$$\Delta\,k_{\|}(\omega_S, \vartheta_S,\, \vartheta_P,\, \vartheta_R) = 0,$$

$$\Delta\,k_{\perp}(\omega_S, \vartheta_S,\, \vartheta_P,\, \vartheta_R) = 0. \qquad (2)$$

Here, the variable ϑ denotes the angular coordinate θ or Φ. Namely, $\vartheta = \theta$ for the wave interaction in the crystallographic planes XZ , YZ and $\vartheta = \Phi$ for the interaction in the XY plane (Fig. 1,*a*). Equations (2) for a fixed ω_S or one of the angles ϑ_i describe (at a given pumping frequency ω_P) the cross sections of the PM surface. These cross sections help us to visualize the characteristic features of the PM surface topology and to develop the classification of the lines and points on this surface that correspond to the noncritical PM of various multiplicities.

The exact conditions of the vector group PM (VGPM) are obtained by the calculation of the full differentials of components of the wave frequency mismatches $\Delta k_{\|}$ and $\Delta k_{\perp}$, Eqs. (2), at $\theta_{P,S} = \text{const}$ with the following nullifying of the determinant of the system of equations linear in $d\omega_S$ and $d\theta_S$. These conditions take the form

$$V_S = V_R \left(\cos\psi - \frac{\sin\psi}{k_R}\frac{\partial k_R}{\partial \theta_R} \right), \qquad (3)$$

where $V_j = \partial\omega_j/\partial k_j = \mathbf{u}_j\mathbf{e}_j \quad (j = S, R)$, $\psi = \theta_{S0} - \theta_{R0}$ is the angle between the directions of propagation of the central spectral components of the signal ω_{S0} and generated ω_{R0} waves, and $\mathbf{u}_j$ are the group speeds of waves ω_j. According to Eq. (3), there is the compensation of a mismatch of the group velocities in the direction of propagation of the signal radiation ω_S under VGPM due to the geometry of the interaction (the term with $\cos\psi$) and the crystal anisotropy. In the main

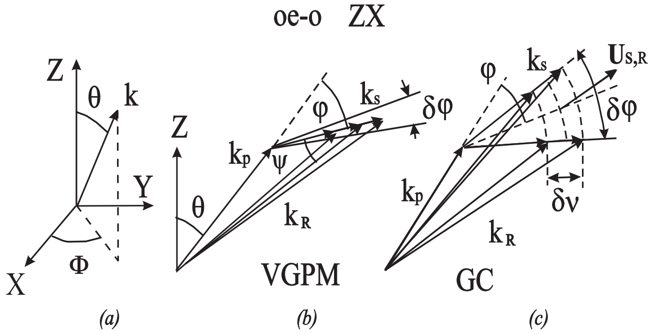

Fig. 1. Diagrams that illustrate the choice of crystallographic axes and the definition of angles that characterize the directions of interacting waves (*a*). Noncritical vector group PM in the frequency of the signal radiation (*b*). Double noncritical PM in the frequency and angles (the group center) (*c*)

planes, where $\partial k_j/\partial\vartheta_j = -k_j\, tg\gamma_j$ (γ_j is the anisotropy angle), condition (3) can be converted to the form

$$V_S = V_R \frac{\cos(\psi - \gamma_R)}{\cos\gamma_R}\,. \qquad (4)$$

The exact VGPM conditions found in [14], which allow the wide-band conversion, are reduced to the equality of the projections of the group speeds of the signal and generated waves with central frequencies ω_{S0} and ω_{R0} on the signal wave propagation directions $\mathbf{k}_S$,

$$\mathbf{u}_S\mathbf{e}_S = \mathbf{u}_R\mathbf{e}_\mathbf{S}\,, \qquad (5)$$

where $\mathbf{e}_S = \mathbf{k}_S/k_S$ is the unit vector in the direction of the signal wave propagation. The similar conditions were obtained in [17]. Schematically, a realization of the VGPM condition in the wide frequency range at fixed θ_S is shown in Fig. 1,*b*. It can be seen that the direction of a signal wave in this case is fixed, $\varphi = \theta_{S0} - \theta_{P0} = \text{const}$. In order to find other noncritical PM conditions, we will derive the relation between changes in the independent variables ω_S, θ_S, and θ_P on a PM surface. In view of the geometry of the interaction shown in Fig. 1,*c*, the system of equations (2) can be written in the form

$$\Delta k_{\|} = k_R - k_S\cos\psi - k_P\cos\beta = 0,$$

$$\Delta k_{\perp} = k_S\sin\psi - k_P\sin\beta = 0, \qquad (6)$$

where $\beta = \theta_{R0} - \theta_{P0}$.

Calculating the full differentials for the longitudinal $\Delta k_{||}$ and transverse $\Delta k_{\perp}$ components of the wave mismatch from Eqs. (6), we obtain, after some transformations, the system of equations

$$\left(\frac{1}{V_R}-\frac{\cos\psi}{V_S}\right)d\omega_S+k_S\,\frac{\sin(\psi+\gamma_S)}{\cos\gamma_S}\,d\theta_S-$$

$$-k_P\,\frac{\sin(\beta-\gamma_P)}{\cos\gamma_P}\,d\theta_P-k_R\,\mathrm{tg}\gamma_R\,d\theta_R=0,$$

$$\frac{\sin\psi}{V_S}d\omega_S+k_S\,\frac{\cos(\psi+\gamma_S)}{\cos\gamma_S}\,d\theta_S-$$

$$-k_P\,\frac{\cos(\beta-\gamma_P)}{\cos\gamma_P}\,d\theta_P-k_R\,d\theta_R=0. \qquad (7)$$

Excluding $d\theta_R$ from Eq. (7), we obtain the relation

$$\Omega\,d\omega_S-G\,d\theta_P+Q\,d\theta_S=0\quad, \qquad (8)$$

where

$$\Omega=1/V_R-\eta_R/V_S,$$

$$G=k_P\sin\left(\beta+\gamma_R-\gamma_P\right)/(\cos\gamma_P\cos\gamma_R),$$

$$Q=k_S\sin\left(\psi+\gamma_S-\gamma_R\right)/(\cos\gamma_S\cos\gamma_R),$$

$$\eta_R=\cos\left(\psi-\gamma_R\right)/\cos\gamma_R.$$

It is seen from condition (8) that, for the vanishing coefficients Ω, G, or Q, each of the independent variables ω_S, θ_S, and θ_P can change in a wide range, which provides the noncriticality of a PM. The condition $\Omega=\Omega_0=0$ corresponds to the VGPM ($d\theta_S/d\omega_S=d\varphi/d\omega_S=0$ at $\theta_P=\mathrm{const}$). The conditions $Q=Q_0=0$ and $G=G_0=0$ correspond to the tangential PM in the signal (TPM; $d\omega_S/d\theta_S=d\omega_S/d\varphi=0$ at $\theta_P=\mathrm{const}$) and in the pumping (TPM; $d\omega_S/d\theta_P=0$ at $\varphi=$ const). The subscript 0 denotes that all the values that determine Ω and Q should be taken for the parameters that correspond to the central frequencies or directions at which the corresponding type of a noncritical PM is realized.

3. Noncritical PM and Group Centers for the Summary Frequency Generation in a Biaxial KTP Crystal

KTP crystals with the transparency band 0.35—4.5 μm are the most promising nonlinear materials. They have the large nonlinear coefficients ($\sim 7\times10^{-12}$ m/V) and the high threshold of an optical damage ($\sim 5\times10^8$ W/cm^2) [18—20]. The angle between the optical axes in a KTP crystal is equal to $2V=34.6°$ (at $\lambda=1.064$ μm) [21]. The optical axes are situated in the XZ plane, therefore the wave interaction in this plane is the most interesting. Consider the conditions for the realization of noncritical PMs in the generation of the summary frequency in the KTP crystal. Since the KTP crystal is positive ($n_e>n_o$) in the XZ plane at $V<\theta\le 90°$, the generated radiation should be an ordinary wave. This is necessary for the compensation of the positive dispersion of the crystal by its anisotropy under the fulfillment of the PM conditions. Thus, the following types of interaction are allowed in the XZ plane: oe—o, eo—o, and ee—o. Analogous types of interaction are possible also in the YZ plane at arbitrary values of θ_P. At $0\le\theta<V$, we have $n_e<n_o$ in the XZ plane. Thus, the KTP crystal is negative, and the sum frequency wave should be a e-wave. In this work, all the calculations were carried out numerically using the dispersion relations [19]

$$n_x^2=2,9971+\frac{0,041030}{\lambda^2-0,038368}-0,012568\,\lambda^2,$$

$$n_y^2=3,0197+\frac{0,044090\,\lambda^2}{\lambda^2-0,042035}-0,012046\,\lambda^2,$$

$$n_z^2=3,3055+\frac{0,063289}{\lambda^2-0,044783}-0,013987\,\lambda^2. \qquad (9)$$

Fig. 2,*a,b* shows angles θ_P and φ versus the signal wavelength λ_S in the whole range of the crystal transparency band, for which it is possible to realize VGPM for the discrete wavelengths of pumping waves.

It can be seen from the figure that, in order to obtain a wide-band vector PM in the KTP crystal as in most other nonlinear crystals, a long wavelength pumping ($\lambda_P>1$ μm) is required, for example, the 1.337- and 1.833-μm radiation of an YAG laser [22]. According to Fig. 2,*a,b*, the VGPM in the XZ plane is realized in the near IR range for the oe—o interaction and in the far IR range for the eo—o interaction. In total, these two interaction types (oe—o and eo—o) allow the tuning of the VGPM almost in the whole transparency range of the KTP crystal except for a small wavelength range 1.81—2.32 μm. On an interaction of the ee—o type in the XZ and YZ planes, we obtain the widest region of the frequency conversion in the VGPM that covers all the transparency band of the KTP crystal (the dashed lines in Fig. 2,*a,b*). This interaction is similar to the oe—o interaction for short wavelengths and to the eo—o interaction for long wavelengths (Fig. 2,*b*). However, in this case, the effective nonlinear coefficient $d_{\text{eff}} = 1/2\,(d_{15} - d_{24}) \sin 2\theta \sin 2\phi$ vanishes at $\phi = 0$ and $\phi = 90°$, that is in the XZ and YZ planes. Therefore, this type of interaction can be realized only for the waves interacting in the intermediate planes that contain the Z axis (for example, at $\phi \approx 45°$). The dashed curves demonstrate the possibility of scanning the VGPM for rays leaving the main dielectric planes of the crystal, where $d_{\text{eff}} \neq 0$. On the curves $\varphi\,(\lambda_S)$ (Fig. 2,*a*), we marked the points that correspond to the conditions for the double noncriticality in the frequency and the divergence angle of the IR radiation ω_S, at which the conditions for the vector group and tangential PMs are fulfilled simultaneously, as shown schematically in Fig. 1,*c*. The horizontal and vertical segments show the frequency and angular intervals, within which the generated radiation intensity decreases twice. This double noncritical PMs in the frequency and one of the angles are called the group center (GC). This center is associated with the minimum of $\theta_P\,(\lambda_S)$ (point A) shown in Fig. 2,*b*. When the geometry of the interaction changes, the wide PM bands shift along the presented VGPM curves. It is obvious that, under the eo—o interaction, the GC is realized only in the collinear case, which is convenient for the wide-band conversion of IR signals and images due to the absence of a walkaff of the signal and resulting radiations. As can be seen from Fig. 2,*b*, the VGPM is possible at two values of λ_S for the oe—o interaction at the fixed angle θ_P, and the fixed wide region $\Delta\lambda_S$ can be converted in frequency at two values of angle θ_P. Smaller values of θ_P on the line $B - A - C - D$ correspond to the case $\varphi > 0$, and greater values of θ_P do to the case $\varphi < 0$, while point B corresponds to the collinear PM ($\varphi = 0$). The minimal value of the signal wavelength at the VGPM is reached at point C (Fig. 2,*b*) at the maximal angle φ (Fig. 2,*a*). The maximal value of λ_S in the VGPM scheme is realized at point B for the collinear interaction. Two branches of the curve $\theta_P\,(\lambda_S)$ are met at point D at $\theta_P = 90°$, where the solutions with $\varphi > 0$ and $\varphi < 0$ become identical. Fig. 2,*a,b* shows that, for both types of the interaction, the size of the PM surface decreases when λ_S approaches ~ 2 μm. It will be shown below that these cases correspond to the shrinking of the PM surface to the critical PM points.

The curves of the wide-band conversion, which are shown in Fig. 2,*a,b*, are the singular lines on the PM

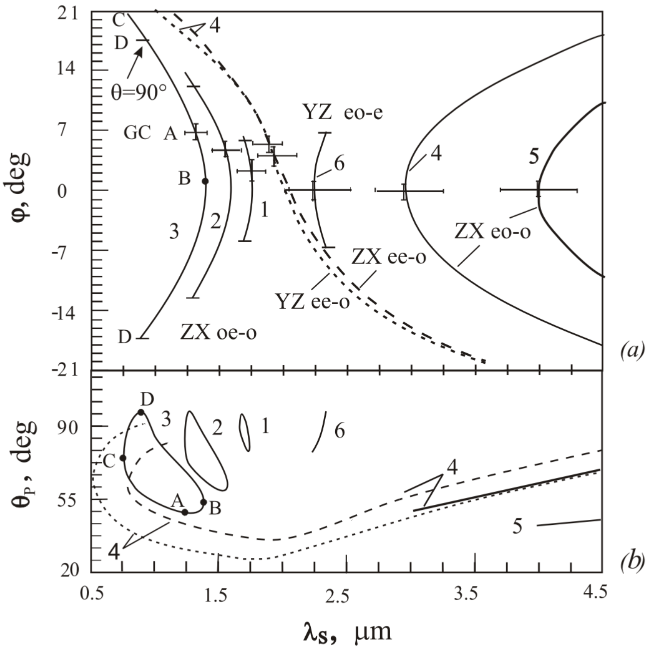

Fig. 2. Functions $\varphi = f\,(\lambda_S)$ (*a*) and $\theta_P = f\,(\lambda_S)$ (*b*) illustrate the possibility of the frequency and angular tunings of the regions of vector group phase matching for a wide-band conversion of the IR radiation for various types of wave interaction in the XZ and YZ planes of a KTP crystal at the pumping wavelengths λ_P: *1* — 1.064 μm, *2* — 1.337, *3* — 1.750, *4* — 1.833, *5* — 1.0795, *6* — 3.8. Horizontal and vertical segments show the spectral and angular widths of the conversion at the group center calculated for the crystal of 1 cm in length

surface in the space of three coordinates λ_S, φ, and θ_P. The projections of this surface at $\lambda_P = 1.064$ μm and the oe—o interaction on the three coordinate planes are shown in Fig. 3,*a,b,c*. In particular, we show the lines of the cross section of the PM surface by planes $\theta_P = \text{const}$ in the coordinates λ_S and φ. This surface has a convex shape, although it can have singularities also (in particular, discontinuities). All possible vector schemes of interactions (including collinear and critical ones) can be associated with this surface. Due to the symmetry of the PM conditions with respect to the change $\theta_P \to 2\pi - \theta_P$, $\varphi \to -\varphi$, Fig. 3 shows only a half of this surface. The VGPM condition is realized along the line that passes through the points of the horizontal tangent lines $d\varphi/\,d\lambda_S = 0$ to these curves (Fig. 3,*a*). It is clear that the top and bottom points of the considered cross sections of the PM surface correspond to two branches of the function $\theta_P\,(\lambda_S)$ shown in Fig. 2,*b*. The wide-band conversion regions can be scanned by the consistent change of θ_P and φ. In contrast to the singular

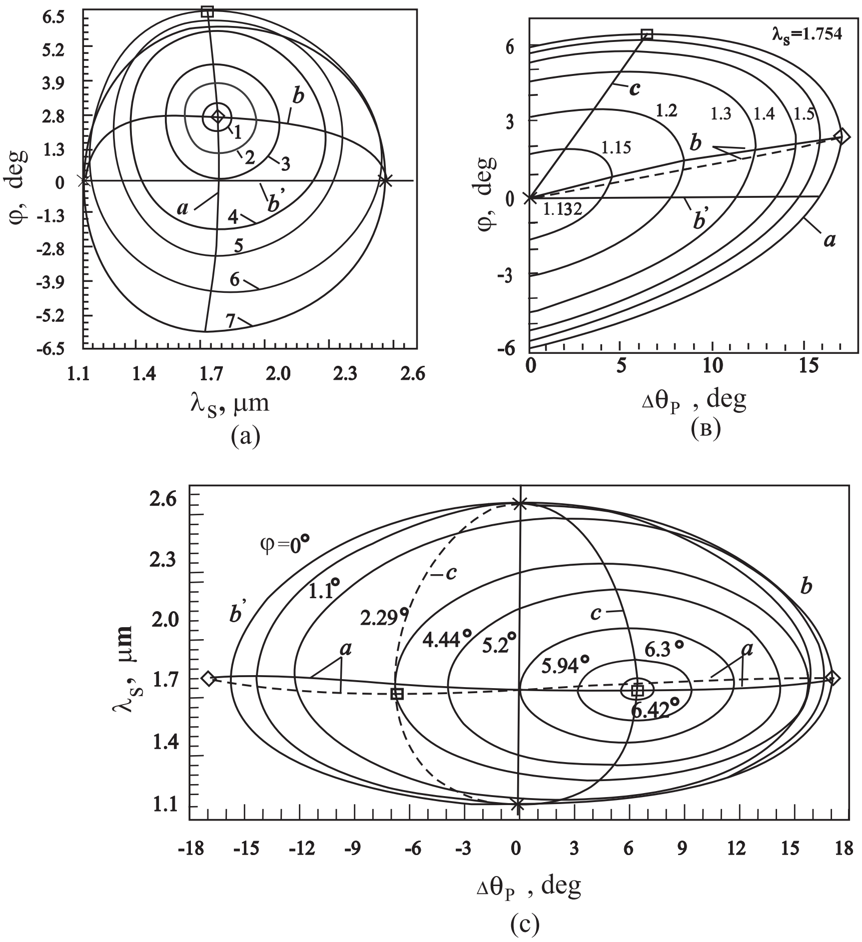

Fig. 3. Cross sections of the PM surface for the oe—o type of interaction in the ZX plane of the KTP crystal at $\lambda_P = 1.064$ μm by planes θ_P = const (a) ($1 - \theta_P = 73.1°$, 2 — 73.5, 3 — 74.25, 4 — 77, 5 — 79, 6 — 83.6, 7 — 90); λ_S = const (b); φ = const (c). Lines a, b, b' and c correspond to the vector group, tangential with respect to the signal and pumping and conditional PM, respectively. Points of the double noncritical PM are marked by the symbols: ◇ — group center, × — tangential center and □ — conditional center

line of the VGPM, most points on the PM surface correspond to narrow-band PMs which can be realized in a wider region of λ_S in comparison with group PMs.

The line connecting the right and left points of the considered cross sections (Fig. 3,a), where $d\varphi/d\lambda_S \to \infty$, corresponds to the noncritical tangential PM (TPM) in the angle φ. At the end points of λ_S on the PM surface, the tangential PM changes to the collinear PM. The tangential PM corresponds to the condition of touching the surfaces of the wave vectors $\mathbf{k}_S$ and $\mathbf{k}_R$ [1, 2] and hence to the same directions of the group speeds of the signal $\mathbf{u}_S$ and generated $\mathbf{u}_R$ radiations. Since the angle between $\mathbf{k}_j$ and $\mathbf{u}_j$ is equal to the anisotropy angle γ_j, the TPM condition $Q = 0$ can be represented in the form $\psi = \gamma_R - \gamma_S$. Due to the weak anisotropy of real crystals, the wave propagation directions at the TPM are close to collinear PMs, which was analyzed in [14] in detail. As can be seen from Fig. 3,a, the angular deviation φ relative to the collinear case in the TPM can reach a few degrees ($\gamma_S = -1,45°$) for the KTP crystal.

The intersection point of two considered lines on the PM surface corresponds to the double noncriticality region, i.e. to the GC. The joint consideration of the conditions of a vector group PM and a collinear PM, which are reduced to the equality of the projections of the group speeds $\mathbf{u}_S$ and $\mathbf{u}_R$ on the direction of the signal propagation and to the requirement of the coincidence of the directions of these speeds, leads to the equality of the group velocities $\mathbf{u}_S = \mathbf{u}_R$ at the GC point. Thus, the GC corresponds to the real group synchronism of waves with double noncriticality in the frequency and the propagation direction of the signal radiation. Fig. 2,b shows that the minimal values of $\theta_{P\,\min}$, at which the GC is realized, decrease, and the PM surface is expanding when λ_S approaches the regions with stronger dispersion. Apart from the tangential PM in respect to the signal, there is also a tangential PM in the pumping which corresponds to the condition $G = 0$ and coincides with the line $\varphi = 0$ for the oe—o interaction (Fig. 3,a). The two lines of the tangential PMs intersect at the extreme left and right points of the PM surface at $\theta_P = 90°$, $\varphi = 0$. The other types of noncritical PMs and the regions of intersection of the lines corresponding to the PM noncritical in one variable, are considered in the next section.

4. Conditional Phase Matchings

For solving a number of applied problems, the promising scheme is that with a double noncritical PM, which is unsensitive to the frequency change and the divergence of one of the waves or to the propagation directions of both waves. For example, the noncritical PM in ω_S and φ can be applied to the conversion of radiation from thermal sources, which allows the visualization of color IR images. Such regions of double noncriticality were revealed from time to time in experimental works [1,9,23—25]. However, we have demonstrated for the first time that GCs exist in all non-linear crystals [14—16].

Using the PM surface, the general classification of noncritical PMs in one variable can be carried out on the basis of the singular lines on this surface. The classification of the points of double noncriticality, which lie on the intersection of these lines, can also be developed. This classification allows a purposeful experimental search for these points. The regions of double noncriticality in the angles φ and θ_P are called tangent centers (TCs). In Fig. 3,a, these centers correspond to the extreme points of the PM surface along the λ_S axis. This type of the double noncritical PM allows us to obtain large angular apertures of the signal and pumping. However, it has a narrow spectral width of the PM. Figure 3,b,c shows the cross sections of the PM surface by the planes $\lambda_S, \varphi = \text{const}$ at the fixed λ_P. These cross sections allow us to consider other types of the double noncritical PM.

In Fig. 2,b, the double noncritical PM in ω_S and φ corresponds to the minimum $\theta_{P\min} = 73.03°$ of the function $\theta_P(\lambda_S)$. Points B and C extremal in λ_S determine the regions of double noncriticality in the angles ω_S and θ_P. Namely, point B corresponds to the wide-band collinear PM tangent in the pumping, while point C corresponds to the maximum with respect to the angle φ on the PM surface. Consider the latter singularity on the PM surface in more details. Note that the maximal value $\varphi_{\max} = 6.43°$ is reached at $\theta_P = 83.57°$. If θ_P further increases, $\varphi_{\max}$ decreases. The same effect takes place in the region $2\pi - \theta_P - \varphi_{\max}$. The cross section of the PM surface by the plane $\theta_P = 90°$ (Fig. 3,a) is symmetric relative to the axis $\varphi = 0$. In the region $\varphi_{\max}$ at a fixed angle between the wave vectors $\mathbf{k}_P$ and $\mathbf{k}_S$, the large changes in the angle θ_P can be observed (Fig. 3,b). Thus, we obtain one more type of the noncritical PM. In this case, the triangle of the wave vectors with a fixed angle φ (Fig. 1), which expresses the momentum conservation, allows the rotations by quite large angles ($2 \div 3°$). Since the large changes in the angles $\theta_{P,S}$ are possible if the condition $\theta_P - \theta_S = \varphi$ is satisfied, we call this type of noncritical PM as a conditional PM (CPM). This type of PM is realized, in particular, for the cylindrical focusing of waves $\omega_{P,S}$. In this case, there is a plenty of pairwise interactions of the radiation components from the wide ranges of the wave vectors directions [6]. The line of a CPM can be found by considering the cross sections of the PM surface by the planes $\lambda_S = \text{const}$ shown in Fig. 3,b. The CPM is realized at the points of the cross sections, where $d\varphi/d\theta_P = 0$. The corresponding line passes in the PM surface from the region $\varphi_{\max}$ to the GC region. In Fig. 3,c, it is line c. Near the intersection point of this curve with the VGPM line at $\theta_P = 83.57°$ and $\varphi = \varphi_{\max} = 6.43°$, we obtain the double noncriticality in ω_S and θ_P which can be called a conditional center (CC). To the best of our knowledge, the CC was not experimentally used till now for the visualization of wide-band IR images.

To obtain the analytical expression for the conditions of the realization of a CPM, we write down Eq. (8) in the form

$$\Omega\, d\omega_S + (Q - G)\, d\theta_P + Q\, d(\theta_S - \theta_P) = 0. \tag{10}$$

At $\omega_S = \text{const}$, this relation and the condition $d\theta_S = d\theta_P$ yield

$$D_0 = Q_0 - G_0 = \frac{k_S}{\cos\gamma_S}\sin(\psi_0 + \gamma_S - \gamma_R) - $$

$$-\frac{k_P}{\cos\gamma_P}\sin(\beta_0 + \gamma_R - \gamma_P) = 0. \tag{11}$$

The last relation should be valid on the PM surface together with (2) or (6). Condition (11) together with the equality $k_P \sin\beta_0 = k_S \sin\psi_0$ can be written as

$$\mathrm{ctg}\psi_0 \sin(\gamma_S - \gamma_R)\cos\gamma_P + ctg\beta_0\,\sin(\gamma_P - \gamma_R)\times$$

$$\times \cos\gamma_S + \sin(\gamma_S - \gamma_P)\sin\gamma_R = 0. \qquad (12)$$

This relation contains only the geometric parameters of the system for central frequencies. Thus, the CPM is realized in the vector scheme of the interaction of waves. Therefore, on the corresponding line, we have $\psi_0 \neq 0$, $\beta_0 \neq 0$ everywhere, except for two points of the double tangential PM ($\varphi = 0$, $\theta_P = 90°$). Consider a CPM with the interaction of two usual and one unusual waves. It follows from Eq. (12) that the unusual wave always propagates at an angle of 90° with respect to the Z axis (along the X axis in the XZ plane). For example, for the oe—o interaction (Fig. 3), the CPM is realized at $\theta_S = \pi/2$ and $\varphi = \pi/2 - \theta_P$. This is related to the appearance of a singularity on the PM surface in the neighborhood of $\varphi_{\max}$. If we define the pumping direction by the deviation $\Delta\theta_P = \pi/2 - \theta_P$ from the XY plane rather than by a deviation from the Z axis, the CPM will be observed along the straight line $\varphi = \Delta\theta_P$. Thus, when the PM surface expands in the direction of the θ_P axis, it also expands regularly in the direction of the φ axis. Below we show that the shrinking of the PM surface in a small region also occurs along all the three coordinates.

5. Frequency Scanning of the Group Centers and a Triple Noncritical PM

As was shown above, there are three closed PM curves on the PM surface that correspond to a noncritical PM in one of the parameters λ_S, φ, or θ_P and also a line of the conditional PM, along which one can observe a large consistent changes of the directions θ_P and θ_S. The intersection points of the first three lines create two pairs of the group centers with the double noncriticality in λ_S and φ (or θ_P) and two tangent centers at the minimal and maximal values of λ_S (the left and right TCs). As was already shown, the line of the conditional PM always passes through a TC, and the intersection of this line with the VGPM line creates a CC. Thus, this region can be considered as the region of a triple noncritical PM (in λ_S, θ_P, and θ_S). A real triple noncritical PM in the variables λ_S, φ, and θ_P will be reached under shrinking the PM surface with changing the pumping wavelength λ_P and with the contraction of all singular lines and points of the double noncritical PM into a small region (a point in the limiting case), after which the PM conditions do not longer hold. The triple noncriticality occurs for each type of the interaction at the exactly determined values of $\lambda_{S,\,P}$. This limiting point can be called a critical point (CP) in view of the violation of the PM conditions. This name is most suitable because this region has properties similar to those in the critical point of matter, in which the difference between the liquid and gaseous states disappears. We suggest that critical points of matter can be related to bifurcations in the structure and properties of matter which occur under the influence of non-linear wave interactions.

In order to scan the central frequency of the conversed wide spectral band in the region of the group or conditional centers, one can change the plane of the wave interaction or the pumping wavelength. The feasibility of the frequency scanning of the VGPM and a change of the PM surface size during a change in the pumping wavelength are illustrated in Fig. 2*a, b*. We have also carried out the detailed investigations of the frequency tuning of the group centers, i.e. of the regions of double noncriticality in λ_S and φ. Figure 4 shows the feasibility of the GC scanning with respect to λ_S under a change in the pumping wavelength λ_P for the waves interacting in the XZ and YZ planes. Along each line of scanning, we have the equality $\mathbf{u}_S = \mathbf{u}_R$ and the noncriticality in the frequency and the signal wave propagation direction in a wide IR and visible ranges. In the same figure, we show the functions θ_P and φ versus λ_S for each GC. It should be noted that $\theta_P \to 0$ in the XZ plane with the increase in λ_P and, correspondingly, angle φ rapidly increases for the oe—o interaction in the short wavelength range of λ_S.

As can be seen from Fig. 4, for interactions of the oe—o and eo—o types, the GCs exist in the whole transparency band of the KTP crystal, with the exception of a small range of the central wavelengths of the signal radiation from 1.813 to 2.318 μm (in the ZX plane) and from 1.831 to 2.214 μm (in the YZ plane). The dashed lines show the GC scanning for the ee—o interaction, for which a non-zero value of $d_{\rm eff}$ is realized when the interaction plane deviates from the XZ and YZ planes. The interaction of this type can provide a smooth GC tuning within the whole transparency band of the KTP crystal. During the GC scanning, other regions of the double noncritical PM also shift.

With decrease in λ_P in the near IR in the case of the oe—o interaction, the PM surface reduces in size, $\theta_P \to 90°$, and $\varphi \to 0$. In a limit at $\lambda_P = 0.9994$ μm, $\lambda_{S0} = 1.813$ μm (the XZ plane) and at $\lambda_P = 0.906$ μm, $\lambda_{S0} = 1.831$ μm (the YZ plane), this surface shrinks into a point, as shown in Fig. 4,*a,b*. The neighborhoods of these points correspond to the triple noncritical PMs in ω_S and two angles φ and θ_P. When approaching this point, the vector PM transfers to collinear PMs ($\varphi = 0$) in the directions of the X and Y axes. The lines of the

GC tuning end in the characteristic CPs, after which the PM conditions do not longer hold ($k_P + k_S < k_R$).

A tendency of the contraction of the PM surface to a CP is also seen in Fig. 2,*a,b*. Analogous CPs are realized under the eo—o interaction in the XZ and YZ planes. The parameters of these CPs are presented in Table 1. In the case of the ee—o interaction in the transparency band of the KTP crystal, the angle θ_P does not approach 90°. Therefore, the region of triple noncriticality is not realized.

For the regions of double noncriticality at $\lambda_P = 1.064$ μm and the oe—o interaction in the XZ plane, we have calculated the signal wavelengths, angles θ_P and φ, and the spectral $\delta\,\nu_S$ and angular $\delta\,\varphi$, $\delta\,\theta_P$ conversion widths which are presented in Table 2. The calculations of $\delta\,\nu_S$ and $\delta\,\varphi$ were carried out with the use of formulas derived in [14] with the assumption that the PM factor $F(\Delta k)$ takes the form $F(\Delta k) = \mathrm{sinc}^2(\Delta k\ l/2)$, where $\mathrm{sinc}\,\tau = \sin\tau/\tau$.

It can be seen from Table 2 that, with the use of a VGPM, we can increase the spectral interval of the converted IR radiation by more than an order and make this interval larger than 500 cm^{-1}. With the use of crystals thinner than 1 cm, we can obtain the spectral conversion widths larger than 1000 cm^{-1}. The double noncriticality of the GC allows us to simultaneously and significantly increase the angular aperture of the signal radiation and to improve the efficiency of conversion of the radiation from heat sources. A large angular divergence of the converted IR signal is admissible for the CC when using a laser radiation with cylindrical divergence in the range of several degrees. We note that the positions of the noncriticality points calculated using other available data on the dispersion of the KTP crystal [26,27] are in good agreement between one another in the near IR range and differ (up to 10 %) in the regions near the boundaries of the absorption bands of the crystal.

T a b l e 1. Laser pumping and IR signal wavelengths, at which the triple noncritical PMs (the critical points of the non-linear interaction) are realized in the KTP crystal

Plane	oe—o interaction		eo—o interaction	
	λ_P, μm	λ_S, μm	λ_P, μm	λ_S, μm
XZ	0.9994	1.813	3.830	2.318
YZ	0.906	1.831	4.048	2.214

T a b l e 2. Spectral and angular conversion widths in the regions of a double noncritical PM on the summary frequency generation in the KTP crystal ($\lambda_P = 1.064$ μm, the oe—o interaction in the XZ plane)

Type of double noncriticality	λ_S, μm	ϕ, deg.	θ_P, deg.	$\sqrt{L}\delta\nu$, cm$^{-1/2}$	$\sqrt{L}\delta\phi$, cm$^{1/2}$deg.	$\sqrt{L}\delta\theta_P$, cm$^{1/2}$deg.
GC	1.754	2.37	73.03	513.5	1.27	
CC	1.693	6.43	83.57	532.4	0.11	3.22
TC (left)	1.132	0	90	43.8	1.09	2.59
TC (right)	2.611	0	90	36.8	1.45	4.04

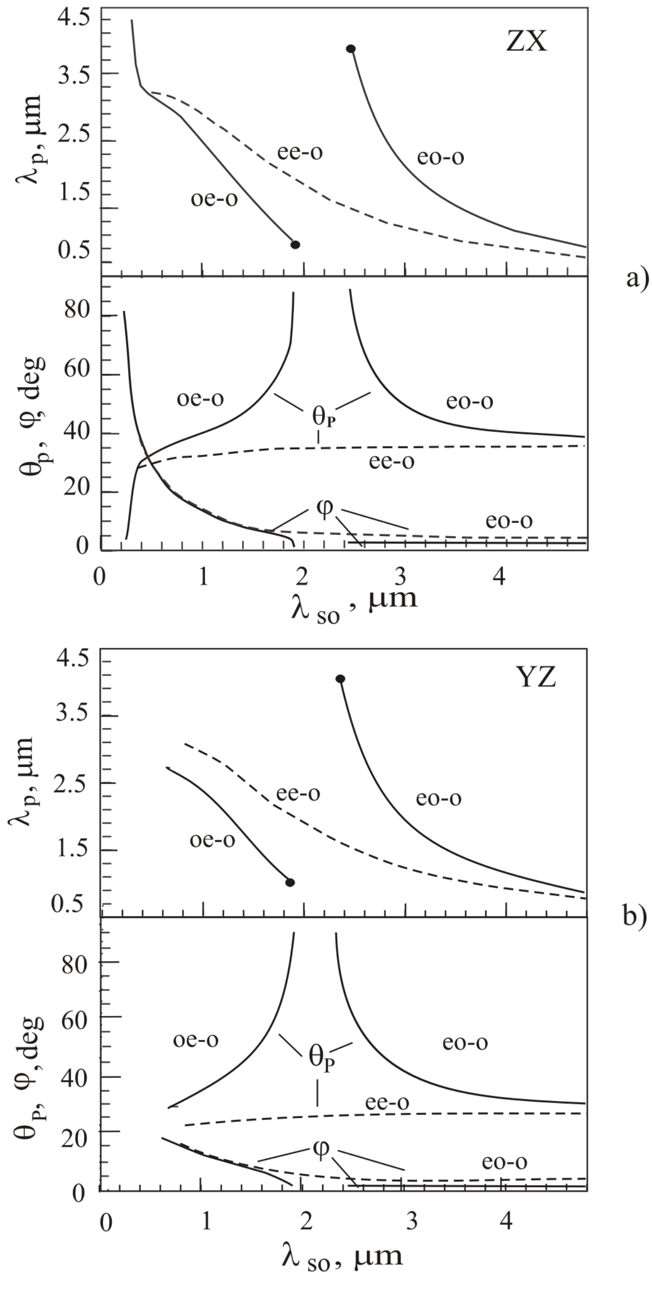

Fig. 4. Scanning the group centers for the summary frequency generation on the vector interactions of various types: in the ZX plane (*a*) and in the YZ plane (*b*) in the KTP crystal. The circles mark the limiting points of a triple noncritical PM

6. Conclusions

In the present work, using the KTP crystal as an example, we have developed a general approach to the analysis of the conditions for the multiple noncritical PM (in the parameters ω_S, θ_S, and θ_P) in biaxial crystals on the summary frequency generation. This approach is based on the analysis of the topology of the PM surface in the space of the frequency and angular coordinates and on the properties of the singular lines on this surface and their intersection points. The wide-band character of the signal radiation and the possibility of the frequency scanning of the monochromatic laser pumping are taken into account. The following results have been obtained.

It is shown that, during the non-linear frequency conversion in biaxial crystals, a noncritical PM in the signal frequency ω_S or in the propagation directions of the signal and pumping waves $\theta_{S,\,P}$ can be realized on the closed lines on the PM surface. In the regions near the intersection points of these lines, the conditions for a double noncritical PM in the frequency ω_S and the angle θ_S (θ_P) or in both these angles are realized.

It is established that, as in the case of uniaxial crystals [14], the equality of the projections of the group speeds of the IR signal and the summary frequency radiation on the propagation direction of the IR radiation is fulfilled in the case of the vector group PM. On the intersection of the lines of the vector group PM and the tangential PM at the group center, the group speed of the signal is equal to that of the generated radiation, $\mathbf{u}_S = \mathbf{u}_R$. For the KTP crystal, the calculations of the spectral $\delta\nu$ and angular $\delta\,\varphi$ conversion widths for the group center have been carried out for a pumping neodymium laser at 1.064 μm. By using crystals of $1 \div 10$ mm in thickness, it is possible to obtain $\delta\nu \approx 500 \div 1500$ cm^{-1} and $\delta\,\varphi \approx 1.5 \div 10°$.

The possibility of the realization of a specific scheme of the angular noncriticality (a conditional PM) has been considered, when the wave propagation directions ($\theta_{P,S}$) are changed simultaneously if the condition $\theta_P - \theta_S = \varphi$ is satisfied. It is shown that the angle φ increases linearly if θ_P deviates from 90°.

The simultaneous realization of a vector group PM and a conditional PM in the region of maximally possible angles $\varphi_{\max}$ between the directions of interacting waves has been demonstrated. This interaction scheme and also the use of GCs allow the conversion of not only wide-band IR signals, but also IR images.

It has been shown that, on the laser wavelength change in the long-wave region $\lambda_P > 1$ μm, the group centers can be scanned in the range $0.7 \div 1.81$ μm in the case of the oe—o interaction and in the range $2.32 \div 4.5$ μm for the eo—o interaction. Under the ee—o interaction on the escape from the XZ or YZ planes, the regions of double noncriticality are scanned in the whole transparency band of the KTP crystal.

The PM critical points are realized at the end points of the lines of the frequency scanning of group centers, when the PM surface shrinks as θ_P approaches 90° (the collinear interaction). In this case, we obtain the triple noncritical PM in the signal frequency and the angles $\theta_{P,S}$ or $\varphi_{P,S}$. The critical points in the XZ plane are realized at $\lambda_P = 0.9994$ μm, $\lambda_S = 1.813$ μm (the oe—o interaction) and at $\lambda_P = 3.830$ μm, $\lambda_S = 2.318$ μm (the eo—o interaction).

The use of multiple noncritical PMs allows the purposeful separation of nonlinear crystals for new devices of quantum electronics.

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КОНЦЕПЦІЯ КРАТНИХ НЕКРИТИЧНИХ ФАЗОВИХ СИНХРОНІЗМІВ ПРИ НЕЛІНІЙНОМУ ПЕРЕТВОРЕННІ ЧАСТОТИ У ДВОВІСНИХ КРИСТАЛАХ

М.Є. Корнієнко, В.І. Задорожний, Т.С. Сіденко

Р е з ю м е

На прикладі двовісного кристала титанілфосфату калію (КТР) розвинуто загальний підхід і проведено аналіз умов некритичних фазових синхронізмів (ФС) різної кратності для процесу генерації сумарної частоти. Підхід ґрунтується на розгляді загальних властивостей поверхні ФС, побудованої в координатах частот і кутів поширення взаємодіючих хвиль, аналізі їх особливих ліній і точок перетину цих ліній. На поверхні ФС розглянуто лінії некритичності за частотою сигнала або кутами поширення хвиль накачки та сигналу. Запропоновано класифікацію двократно некритичних ФС (за частотою та одному з кутів або за обома кутами). Встановлено, що область двократної некритичності за частотою та розбіжністю ІЧ-сигналу може скануватися по всій області прозорості кристала КТР при перестроюванні частоти довгохвильової лазерної накачки. Проаналізовано новий тип кратного некритичного синхронізму (умовний ФС), який реалізується за узгодженої зміни напрямків поширення накачки та сигналу в області максимальної величини кута між взаємодіючими хвилями. Показано можливість реалізації некритичності за трьома незалежними параметрами — за частотою та двома кутами. Кратні некритичні ФС дозволяють як візуалізовувати широкосмугові ІЧ-спектри та ІЧ-зображення, так і використовувати ці методи при перетворенні частоти фемтосекундних лазерних імпульсів.