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Broadband tunable and multiple noncritical phase matchings in LBO crystal

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The multiple (double and triple) phase matchings (noncritical in the frequency and propagation directions of interacting waves) for the sum frequency generation in a lithium triborate (LBO) crystal have been studied. The use of the vectorial group phase matching and multiple noncritical ones for frequency conversion in a LBO crystal makes it possible to extend substantially the tuning region of the broadband frequency conversion up to overlapping essentially the whole region of the crystal transparency (0.45 to 2.6 μm) and to attain a large conversion band (≥ 2000 cm^{-1}) at a large permissible acceptance angle of the signal radiation (about 2° for about 3 mm thick crystals) within a broad spectral range. The broadband region of frequency conversion is tuned by variation of the pump wavelength within 0.54 to 2.6 μm range using various interaction types in the principal planes: XZ (ee-o, eo-e, oe-e), YZ (eo-o, oe-o), and XY (oo-e). The broadband frequency conversion of IR radiation at noncritical phase matching combined with the large permissible pumping divergence is expected to be useful in the IR image visualization.

Исследованы направлением объемной и интегральной нелинейной оптики. В работе исследуются кратные (двойные и тройные) некритичные (по частоте и направлениям распространения взаимодействующих волн) фазовые синхронизмы в кристалле трибората лития (LBO) для процесса генерации суммарной частоты. Использование векторного группового и кратных некритичных фазовых синхронизмов для преобразования частоты в кристалле LBO позволяет как существенно расширить область перестройки широкополосного преобразования частоты с целью перекрыть практически всю область прозрачности кристалла LBO (0.45–2.6 мкм), так и достичь ширины полосы преобразования (≥ 2000 $см^{-1}$) при большом допустимом угле приема сигнального излучения (~2° для кристаллов толщиной ~3 мм) в широкой спектральной области. Перестройка области широкополосного преобразования частоты осуществляется путем изменения длины волны накачки в диапазоне 0.54–2.6 мкм с использованием различных типов взаимодействия в главных оптических плоскостях: XZ (ee-o, eo-e, oe-e), YZ (eo-o, oe-o) и XY (oo-e). Ожидается, что широкополосное преобразование частоты ИК- излучения в условиях некритичного фазового синхронизма в сочетании с большой допустимой расходимостью накачки будет полезным для визуализации ИК изображений.

In recent years, there is an increasing interest in broadband non-collinear sum- and difference frequency generation due to the progress in the development of new pumping sources, in particular, tunable lasers, parametric frequency converters, and parametric generators of short and ultrashort (up to 10 fs) pulses [1–7]. The three-wave parametric processes tunable in a broad spectral region from UV to mid-IR have a number of important applications to frequency- and time-resolved spectroscopy for the study of fast physical, chemical, and biological processes. The progress in sum-frequency vibrational spectroscopy, pulse-probe-type ultrafast spectroscopy is a convincing example thereto [6]. Among different nonlinear media for frequency up-converters, it is the lithium triborate (LBO) biaxial crystal that is a very promising material because of its high nonlinearity, wide transparency region (from 0.35 to 4.5 μm), high optical quality, and high damage threshold. This crystal will be the object to study in this article.

In our opinion, insufficient attention has been given up to now to the question of utilization of multiple noncritical phase matchings in optical devices. In many cases, the near-90° phase matching (PM) noncritical in phase angle and tangential PM (TPM) were employed. The main drawback of these PM's is a limited tuning range. Many works deal with the collinear and vectorial (noncollinear) group PM (VGPM), where, besides the usual PM conditions

$$\omega_r = \omega_p \pm \omega_s, \quad \mathbf{k_r} = \mathbf{k_p} \pm \mathbf{k_s}, \tag{1}$$

the equality of the projections of the group velocities $\mathbf{V}_s$ and $\mathbf{V}_r$ onto the $\mathbf{k}_r$ direction is required at narrow-band pumping [4–9]. The subscripts "p, s, r" in (1) refer to the pump, signal, and resulting waves, respectively. The use of the noncritical vectorial group PM extends the conversion possibilities, in particular, it makes it possible to scan the broadband group PM region along the crystal transparency region. In addition, the combination of the tangential and vectorial group PM's (double noncritical PM) provides the simultaneous realization of a broadband and wide-angle conversion of signals and images. Using the different pumping wavelengths, the double noncritical PM can be displaced towards shorter or longer wavelengths.

A schematic diagram of the noncollinear wave interaction for sum frequency generation is shown in Fig. 1. Here, θ_i ($i = p$, s, r) is the angle between the wave vector $\mathbf{k}_i$ and the S axis, $\varphi = \theta_s - \theta_p$, $\beta = \theta_r - \theta_p$, $\psi = \varphi - \beta = \theta_s - \theta_r$. Equations (1) define the phase matching surface in the three-dimensional space with coordinates ω_s, θ_p, φ at fixed pump frequency. Projecting the vectorial equation included in (1) onto the $\mathbf{k}_r$ direction and onto perpendicular one, we obtain the system of equations

$$k_p(\theta_p)\cos(\theta_r - \theta_p) + k_s(\omega_s,\theta_s)\cos(\theta_s - \theta_r) = \\ = k_r(\omega_r,\theta_r),$$

$$k_p(\theta_p)\sin(\theta_r - \theta_p) - k_s(\omega_s,\theta_s)\sin(\theta_s - \theta_r) = 0. \tag{2}$$

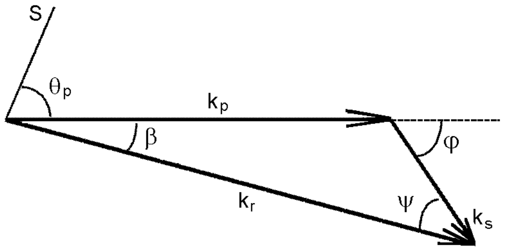

Fig. 1. Geometry of noncollinear wave interaction in a LBO crystal. S is the principal Z axis for the interaction in the XZ and YZ planes of the optical frame or the principal X axis for the interaction in the XY plane.

Taking the total differential with respect to ω_s, θ_p, θ_s, and θ_r, after eliminating the variable $d\theta_r$ we obtain the following equation relating the changes in ω_s, θ_p, θ_s on the phase matching surface

$$A\, d\omega_s - P\, d\theta_P + Q\, d\theta_s = 0, \tag{3}$$

where $A = \cos\gamma_r/V_r - \cos(\psi_0 - \gamma_r)/V_s$, $P = k_p\sin(\beta_0 + \gamma_r - \gamma_p)/\cos\gamma_p$, $Q = k_s\sin(\psi_0 + \gamma_s - \gamma_r)/\cos\gamma_s$, $\gamma_i = -1/\tan((1/k_i)\partial k_i/\ \partial\theta_i)$ are the Poynting vector walk-off angles; $V_i = \partial\omega_i/\partial k_i$ are the projections of the corresponding group velocities $\mathbf{V}_i$ onto the $\mathbf{k}_i$ directions. The subscript "o" refers to the central frequencies and angles of interacting waves. The condition $A = 0$ corresponds to the equal projections of the group velocities $\mathbf{V}_s$ and $\mathbf{V}_r$ onto the direction of $\mathbf{k}_s(\omega_{s0}, \varphi_0)$ and determines the broadband vectorial group phase matching (VGPM). The conditions $P = 0$, $Q = 0$ determine the case of tangential phase matching (TPM) corresponding to the collinearity of vectors $\mathbf{V}_p \| \mathbf{V}_r$ and $\mathbf{V}_s \| \mathbf{V}_r$ or tangency of the wave vector surfaces $\mathbf{k}_p$, $\mathbf{k}_r$ and $\mathbf{k}_s$, $\mathbf{k}_r$. Any of the above-mentioned conditions is achieved along closed curves lying in the phase-matching surface and describes a single noncritical PM.

If the absolute values of the group velocities $\mathbf{V}_s$ and $\mathbf{V}_r$ are equal and their directions are the same, a double noncritical PM (DNPM) is achieved. In this case, the phase matching is noncritical both in the signal frequency and in the value of angle φ. This case will be referred to as a "group center" (GC). The physical sense of the above DNPM condition consists in compensation of the group velocity mismatch due to the geometry of interaction and crystal anisotropy. Moreover, there are other DNPM types. The second type is the PM noncritical in both the frequency ω_s and the angle θ_p ($A = P = 0$) and the third type is the PM

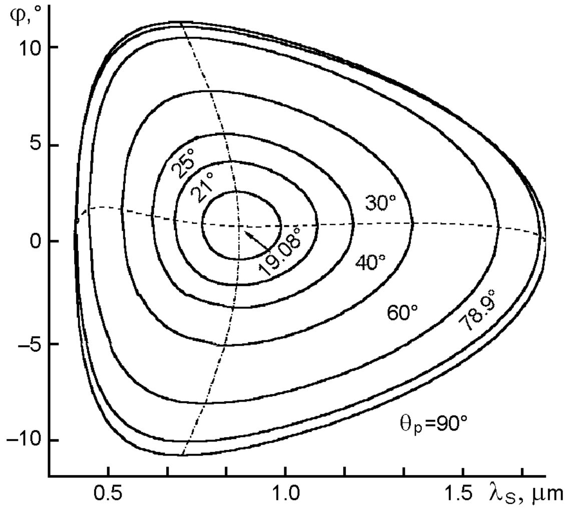

Fig. 2. Sections of the phase matching surface for sum frequency generation (oe-o interaction in the YZ plane) in the LBO crystal pumped at 1.064 μm.

noncritical in angles θ_s and θ_p ($Q = P = 0$). The last case will be referred to as a "tangential center" (TC). The TC is realized as a rule in the case of a collinear interaction. The fourth type of DNPM is called a "conditional double noncritical phase matching" (CDNPM). This PM is noncritical in angles θ_p and θ_s at a fixed angle φ. The condition $Q = P$ ($Q,\ P \neq 0$) for CDNPM can be found after transforming (3) into the form $Ad\omega_s + (Q - P)\ d\theta_p + Qd\varphi = 0$. In contrast to the TC case where PM is noncritical at independent changes in angles θ_p and θ_s, the angles θ_p and θ_s are related to each other by the relation $\theta_p - \theta_s = \varphi$ in the case of CDNPM. The intersection point of the curves in the PM surface where VGPM and CDNPM are realizable simultaneously, will be referred to as a "conditional triple center" (CTC).

There is a interaction geometry of the optical waves where PM is noncritical in the three independent variables ω_s, θ_p, and φ (or θ_s). This PM type is achieved when the PM surface practically degenerates into a point of triple PM. The existence domains of this triple PM in anisotropic biaxial crystals are defined by the anisotropy characteristics and achieved along the principal axes of the optical frame where the difference between the refraction indices (for example, $n_x - n_z$ or $n_y - n_z$) reaches its maximum.

It is well known [3] that biaxial crystals have more opportunities for fulfillment of PM conditions at nonlinear interaction of waves. This occurs because the optical axes do not coincide with the principal axes of the optical frame X, Y, Z.

To consider the existence conditions of noncritical PMs in the LBO crystal at sum frequency generation (SFG), we solved numerically equations (1) for all possible interaction geometries in the planes XZ, XY and YZ. In these calculations, the following Sellmeier equations were used describing the dispersion of refractive indices for a Fujian CASTECH LBO crystal

$$n_x^2 = 2.454140 + \frac{0.011249}{\lambda^2 - 0.011350} - 0.014591\,\lambda^2 - 6.60\cdot 10^{-5}\,\lambda^4, \tag{4}$$

$$n_y^2 = 2.539070 + \frac{0.012711}{\lambda^2 - 0.012523} - 0.018540\,\lambda^2 + 0.20\cdot 10^{-3}\,\lambda^4,$$

$$n_z^2 = 2.586179 + \frac{0.013099}{\lambda^2 - 0.011893} - 0.017968\,\lambda^2 - 2.26\cdot 10^{-4}\,\lambda^4,$$

where λ is the wavelength in μm.

The calculated sections of the PM surface by planes $\theta_p = const$ for sum frequency generation in a LBO crystal pumped at 1.064 μm are shown in Fig. 2. The dash-point-line shows the extreme points of the PM curves where $d\varphi/d\lambda_s = 0$ and broadband (noncritical in ω_s) vectorial group phase matching (VGPM) occurs. The dashed line shows the extreme points of the PM curves in which $d\varphi/d\lambda_s \to \infty$ and tangential phase matching (TPM), admitting a large angular aperture of the signal wave, is achieved.

As is seen from Fig. 2, the collinear group PM ($\varphi = \beta = 0°$) occurs at $\theta_p = 19.44°$ and $\lambda_s = 944.5$ nm. At $\theta_p = 90°$, the TPM lines for signal and pumping radiation are crossed at $\lambda_s = 385$ nm and 1.982 μm forming two "tangential centers" (TC) (double PM noncritical in angles). A double PM noncritical in both the frequency and the angle ("group center", GC) is realized at minimum angle $\theta_p = 19.08°$ for $\lambda_s = 943.6$ nm, $\varphi = 0.73°$, $\beta = 0.39°$. A triple PM noncritical in ω_s and dependent variables θ_s and θ_p ("conditional center", CDNPM), takes place at $\lambda_s = 743.7$ nm, $\varphi = 11.19°$, $\beta = 6.69°$, $\theta_p = 78.85°$.

Typical dependences of spectral bandwidth $\delta\nu$ and angular width (acceptance angle) $\delta\varphi$ of signal radiation on signal wavelength λ_s for a 3-mm-long LBO crystal cal-

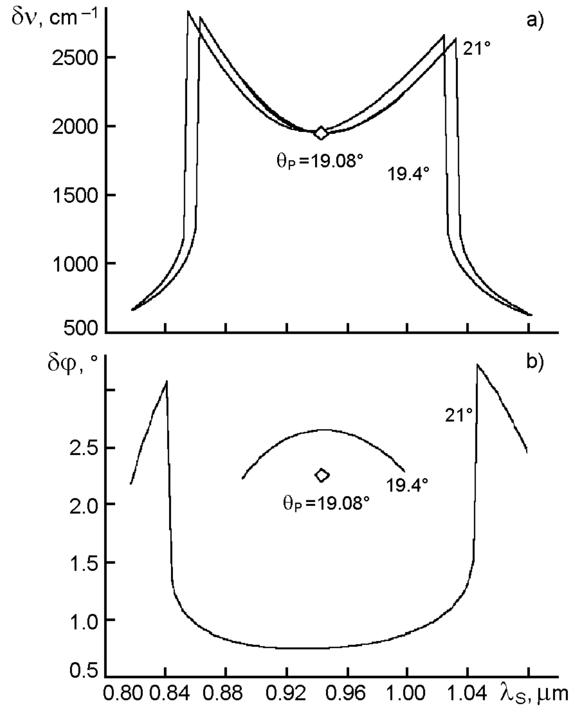

Fig. 3. Spectral bandwidth (a) and acceptance angle (b) of signal radiation as functions of the signal wavelength for a 3 mm long LBO crystal pumped at 1.064 μm near the DNPM region (oe-o interaction in the YZ plane).

culated at the half-intensity level of generated radiation by the method proposed in [8] are shown in Fig. 3(a) and Fig. 3(b), respectively. These figures show that the use of the noncritical phase matchings mentioned above improve the characteristics of converters, in particular, increase the magnitude of the conversion band up to ~$2.5 \cdot 10^3$ cm^{-1} and acceptance angle up to 2.3°.

The tuning curves for the double noncritical phase-matching regions where both a large phase-matching bandwidth and a large acceptance angle of signal radiation occur are realized are shown in Fig. 4. Referring to this Figure, the double noncritical PMs in a LBO crystal is achieved in the wide spectral region from 0.45 to 2.6 μm when changing the pump wavelength from 0.54 μm to 2.6 μm. When λ_p changes towards shorter wavelengths for eo-e interaction in the XZ plane, the PM surface becomes smaller and reduces into a triple noncritical PM point at $\lambda_p = 1.13$ μm ($\lambda_s = 916$ nm). There are also two points of triple noncritical PM for the oe-o interaction in the YZ plane at $\lambda_p = 1.13$ μm ($\lambda_s = 916$ nm) and $\lambda_p = 762$ nm, ($\lambda_s = 1.098$ μm). In all cases of the triple

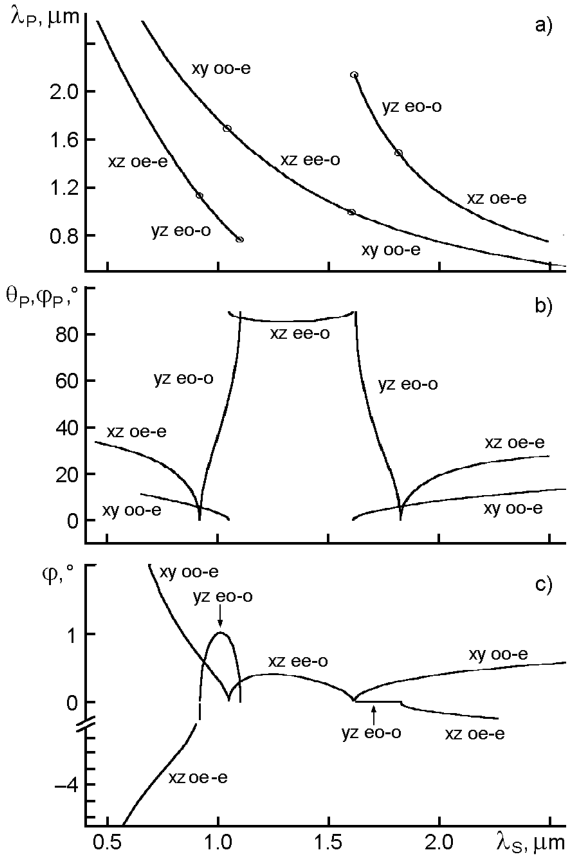

Fig. 4. Pump wavelength (a), pump angles (θ_p in the XZ and YZ planes and φ_p in the XY plane) (b) and angle φ (c) as functions of the signal wavelength for all possible interaction types in principal optical planes of LBO crystal for sum frequency generation under conditions of multiple phase matchings. The double phase matching noncritical in λ_s and φ is realizable along solid curves. Circles are the points where a triple noncritical phase matchings are attained.

noncritical phase matching, VGPM becomes converted into a collinear PM.

In a similar way, as λ_p changes towards longer wavelengths for oe-e interaction in the XZ plane, the PM surface becomes smaller and reduces into a triple noncritical PM point at $\lambda_p = 1.482$ μm ($\lambda_s = 1.823$ μm). Two triple noncritical PM points where the PM surface degenerates into a point exist for eo-o interaction in the YZ plane. These PM's are realized at $\lambda_p = 1.482$ μm ($\lambda_s = 1.823$ μm) and $\lambda_p = 2.137$ μm ($\lambda_s = 1.621$ μm). There are also the triple noncritical PM points at λ_p = 994 nm ($\lambda_s = 1.608$ μm) for the oo-e interaction in the XY plane and $\lambda_p = 1.685$ μm

($\lambda_s = 1.046$ μm) for oe-o interaction in the XZ plane (see Fig. 4(a)) .

The oe-o interaction in the YZ plane is of a particular practical interest. When the LBO crystal is pumped at 1.13 μm, for example, by a Nd:YAG pumped near-IR dye laser and potassium titanyl phosphate (KTP) optical parametric oscillator, a 3-mm-long nonlinear converter permits the conversion of the 0.84–1.01 μm spectral region into a visible one (0.48–0.53 μm) at the acceptance angle for signal radiation about 2.3° and maximum allowable pumping divergence of 11°. Other triple noncritical PM points are of a lesser practical interest for lack of suitable pumping sources.

To conclude, we have investigated different possible types of noncritical phase matchings in a LBO crystal. We have shown that along with well known single noncritical phase matchings, there exist double and triple ones in this crystal. The double PM noncritical in the wavelength and angle of signal radiation in the LBO crystal occurs at different types of interactions in all optical planes XZ (ee-o, eo-e, oe-e), YZ (eo-o, oe-o) and XY (oo-e) and can be tuned within 0.45–2.6 μm spectral region by changing the pumping wavelength from 0.54 to 2.6 μm. In order to realize the double noncritical PM of "group center" type, long-wave lasers can be used: Nd^{3+} ones using different solid-state matrices and emitting at 1.33–1.37 μm and 1.833 μm, as well as solid-state lasers utilizing activating ions Ho^{3+}, Tu^{3+} and Dy^{3+} that generate at 2.0654, 2.348 and 2.358–2.366 μm, respectively. Besides, we have found six points of triple (noncritical in pump angle, signal wavelength and signal angle) PM in the LBO crystal. The triple noncritical PM point at pump wavelength of 1.13 μm and signal wavelength of 916 nm is of a particular practical interest. In this case, a 3-mm long LBO-based nonlinear converter permits the broadband conversion ($\delta\nu \sim 2027$ cm^{-1}) of 0.84–1.01 μm spectral region into visible one (0.48–0.53 μm) at the acceptance angle $\delta\varphi \sim 2.3°$ of the signal radiation and maximum allowable pumping divergence of about 11°. The broadband conversion of IR radiation under these conditions is expected to be useful in visualization of IR images.

Thus, we have shown that the use of multiple noncritical PM's in up-converters increases the bandwidth and angular acceptance for signal radiation, practically eliminates the Poynting vector walk-off and, consequently, increases the conversion efficiency and rises the parametric gain, thereby providing a tool for fast nonlinear conversion of signals and IR images into visible ones. Besides, the use of the above-mentioned noncritical PM improves the characteristics of available light sources and converters taking advantage of the high stability of the devices against disturbances.

References

1. Chin-Wei Hsu, C.C.Yang, *Optics Lett.*, **26**, 1412 (2001).
2. A.Dubietis, G.Tamoauskas, A.Varanaviius et al., *Optics Lett.*, **25**, 1116 (2000).
3. V.I.Zadorozhnii, *Optics Commun.*, **176**, 489 (2000).
4. A.Shirakawa, T.Kobayashi, *Appl. Phys. Lett.*, **72**, 147 (1998).
5. V.Krylov, J.Gallus, U.P.Wild et al., *Appl. Phys. B*, **70**, 163 (2000).
6. T.Kobayashi, A.Baltuska, *Meas. Sci. Technol.*, **13**, 1671 (2002).
7. J.Wang, M.H.Dunn, C.F.Rae, *Optics Lett.*, **22**, 763 (1997).
8. S.G.Dolinchuk, N.E.Kornienko, V.I.Zadorozhnii, *Infrared Phys. Technol.*, **35**, 895 (1994).
9. G.M.Gale, M.Cavallari, T.J.Driscoll et al., *Optics Lett.*, **20**, 1562 (1995).

Широкосмугові та кратні некритичні фазові синхронізми у кристалі LBO

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Досліджені кратні (подвійні та потрійні) некритичні (за частотою та напрямкам поширення взаємодіючих хвиль) фазові синхронізми у кристалі трибората літію (LBO) для процесу генерації сумарної частоти. Використання векторного групового і кратних некритичних фазових синхронізмів для перетворення частоти у кристалі LBO дозволяє як істотно розширити область перестроювання широкосмугового перетворення частоти з метою перекриття практично всієї області прозорості кристала LBO (0.45–2.6 мкм), так і досягти ширини смуги перетворення (≥ 2000 см$^{-1}$) при великому припустимому куті прийому сигнального випромінювання (~2° для кристалів товщиною ~3 мм) у широкій спектральній області. Перестроювання області широкосмугового перетворення частоти здійснюється шляхом зміни довжини хвилі накачування у діапазоні 0.54–2.6 мкм із використанням різних типів взаємодії у головних оптичних площинах: XZ (ee-o, eo-e, oe-e), YZ (eo-o, oe-o) та XY (oo-e). Очікується, що широкосмугове перетворення частоти ІЧ-випромінювання в умовах некритичного фазового синхронізму у сполученні з великою припустимою розбіжністю накачування буде корисним для візуалізації ІЧ-зображень.