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Temperature-noncritical third harmonic generation in an LBO crystal

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Abstract. **Temperature-noncritical third harmonic generation is studied theoretically and experimentally in an LBO crystal by compensating for the temperature-induced deviation of the phase-matching direction due to thermal strains in the crystal. The width of the phase-matching temperature curve obtained experimentally in a 14.1-mm-long LBO crystal for the third harmonic generation of the 1.064-μm radiation is 73 °C.**

Keywords: *third harmonic generation, frequency conversion, phase matching, phase-matching temperature width, thermal-strain-induced changes.*

The possibility of realising temperature-noncritical interactions of waves during laser-radiation frequency conversion is of obvious scientific and practical interest. A frequency conversion is usually rather critical to changes in the crystal temperature. This feature manifests itself in a strong temperature dependence of the wave detuning $\Delta k(T)$, which, in turn, is determined by a significant effect of thermal actions and self-actions [1, 2]. As a result, during the generation of, for example, the second (SHG) and third (THG) harmonics, as well the sum and difference frequencies, a nonlinear crystal must be temperature-stabilised. However, this cannot fully exclude a strong negative effect of thermal self-actions due to the presence of thermal inhomogeneities over the crystal cross section. The solution to this problem depends on the possibility of achiving temperature-noncritical frequency-conversion regimes, for which at least the first temperature derivative of the wave detuning is zero ($\mathrm{d}\Delta k/\mathrm{d}T = 0$). In this case, the finite width of the phase-matching temperature curve (the so-called temperature width) ΔT_{pm} is determined by the second and higher temperature derivatives of the wave detuning.

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The possibility of achieving the temperature-noncritical SHG in a KTP crystal was demonstrated theoretically and experimentally in [3, 4]. The width of the phase-matching temperature curve obtained experimentally in a 7.7-mm-long KTP crystal for the sff interaction was $\Delta T_{\mathrm{pm}} = 210\,^{\circ}\mathrm{C}$.

A violation of phase matching upon temperature changes is generally determined by the action of three mechanisms: temperature-induced changes in the refractive indices, thermal strains in crystals, and a violation of the interaction collinearity (see, e.g., Refs [5, 6]). A certain role can also be played by elastooptical changes in the refractive indices indirectly associated with thermal effects [7]. Note that, although the effect of direct temperature-induced changes in the refractive indices was studied in detail [1], thermal-strain-induced changes in a crystal as a whole, which manifest themselves, e.g., as rotations of crystal faces caused by an anisotropy of its linear expansion coefficients, have been studied inadequately.

It was shown in Refs [5, 6] that, in crystals having large linear expansion coefficients with a significant anisotropy, the phase-matching temperature width during frequency conversion largely depends on the crystal mounting, which is determined by the effect of thermal-strain-induced changes. The widths (FWHM) of the phase-matching temperature curve obtained experimentally for $\mathrm{YAG:Nd^{3+}}$ laser radiation (1.064.2 nm + 532.1 nm) in a 5.33-mm-long LBO crystal for the ssf interaction in the xy plane during the THG were 32.2 and 10.6 °C, when the crystal was pressed to the holder base with its lateral and entrance faces, respectively. These results correspond to the temperature-critical phase matching ($\mathrm{d}\Delta k/\mathrm{d}T \neq 0$).

This paper continues the studies performed in Refs [5, 6] by investigating the possibility of achieving a temperature-noncritical THG for $\mathrm{Nd^{3+}:YAG}$ laser radiation (1.064.2 nm + 532.1 nm) in an LBO crystal and using the mechanism of thermal-strain-induced changes in the crystal at various methods of its mounting for compensating for the deviation of the phase-matching direction caused by temperature-induced changes in the refractive indices. A collinearity violation and effects of elastooptical changes in the refractive indices in the LBO crystal are small and can be ignored [6].

When the crystal is heated, the temperature-induced changes in the refractive indices lead to a change in the phase-matching direction, which is determined by the angle with respect to the x axis, in the xy plane ($\theta_{\mathrm{pm}} = 90^{\circ}$); in this case, $\mathrm{d}\varphi_{\mathrm{pm}}/\mathrm{d}T = -0.155$ ang. min $^{\circ}\mathrm{C}^{-1}$.

The change in the direction of the radiation propagation, caused by the effect of thermal-strain-induced changes in crystal faces, depends on the crystal mounting. In the problem considered, when the crystal entrance or lateral face is pressed to the holder base, the deviation of the radiation direction from the initial direction inside the crystal is determined by the derivative $\mathrm{d}\Delta\varphi^{(1)}/\mathrm{d}T = +0.3316$ ang. min °C^{-1} or the derivative $\mathrm{d}\Delta\varphi^{(2)}/\mathrm{d}T = -0.0796$ ang. min °C^{-1}, respectively.

The total angular detuning $\Delta\varphi_s$ is determined by the angular difference of the phase-matching and radiation-propagation directions. Obviously the detuning value depends on the method of the crystal mounting. In the xy plane, the expression for the angular detuning has the form

$$\frac{\mathrm{d}\Delta\varphi_s^{(j)}}{\mathrm{d}T} = \frac{\mathrm{d}\varphi_{\mathrm{pm}}}{\mathrm{d}T} - \frac{\mathrm{d}\Delta\varphi^{(j)}}{\mathrm{d}T}. \tag{1}$$

When the crystal is heated, the thermal-strain-induced rotation of the radiation-propagation direction generally ensures a partial compensation for the rotation of the phase-matching direction, if both terms on the right-hand side of (1) have identical signs. If, in addition, the condition $\mathrm{d}\Delta\varphi^{(j)}/\mathrm{d}T = \mathrm{d}\varphi_{\mathrm{pm}}/\mathrm{d}T$ is satisfied, the temperature-induced change in the phase-matching direction is fully compensated for; i.e., the completely temperature-noncritical phase matching ($\mathrm{d}\Delta k/\mathrm{d}T = 0$) is observed for all thermal processes in the crystal. However, for the LBO crystal under study, we have $\mathrm{d}\Delta\varphi_s^{(1)}/\mathrm{d}T = +0.487$ ang. min °C^{-1} and $\mathrm{d}\Delta\varphi_s^{(2)}/\mathrm{d}T = +0.0754$ ang. min °C^{-1}; i.e., the temperature-critical phase matching is realised in both versions of crystal mounting. As was shown in Ref. [6], the largest width of the phase-matching temperature curve is attained when the lateral face of the crystal is pressed to the holder base.

All the data presented above referred to the conditions under which both the radiation-propagation and phase-matching directions lie in the xy plane. As was shown in Refs [4, 8], the direction of temperature-noncritical phase matching may lie not only in the principal crystal planes. The directions of temperature-noncritical interactions (these directions are determined only by temperature-induced changes in the refractive indices) in the space of angles φ and θ form fourth-order conical surfaces similar to the phase-matching directions [9]. Figure 1 shows the phase-matching directions (thin lines) and the phase-matching temperature widths (the wide bands with variable blackening densities) for the THG (1.064.2 nm + 532.1 nm) in the LBO crystal for two types of interactions: ssf (Fig. 1a) and fsf (Fig. 1b). The dark region corresponds to the maximum temperature width of 50 °C and more, and the central line in this region corresponds to the infinitely large phase-matching temperature width. In the adjacent regions characterised by different degrees of blackening, the phase-matching temperature widths differ by 10 %.

It follows from these results that, for the ssf interaction in LBO, there is a direction ($\varphi = 39.3°$ and $\theta = 74.8°$) along which these curves intersect. The latter corresponds to the simultaneous presence of both the phase matching and the widest phase-matching temperature curve (Fig. 1a). This is just the temperature-noncritical phase-matching regime ($\mathrm{d}\Delta k/\mathrm{d}T = 0$), which is achieved when the temperature-strain-induced changes in the crystal as a whole can be ignored. The direction found does not lie in the xy plane in which the angular-noncritical phase matching can also be achieved. However, calculations have shown that a wave-

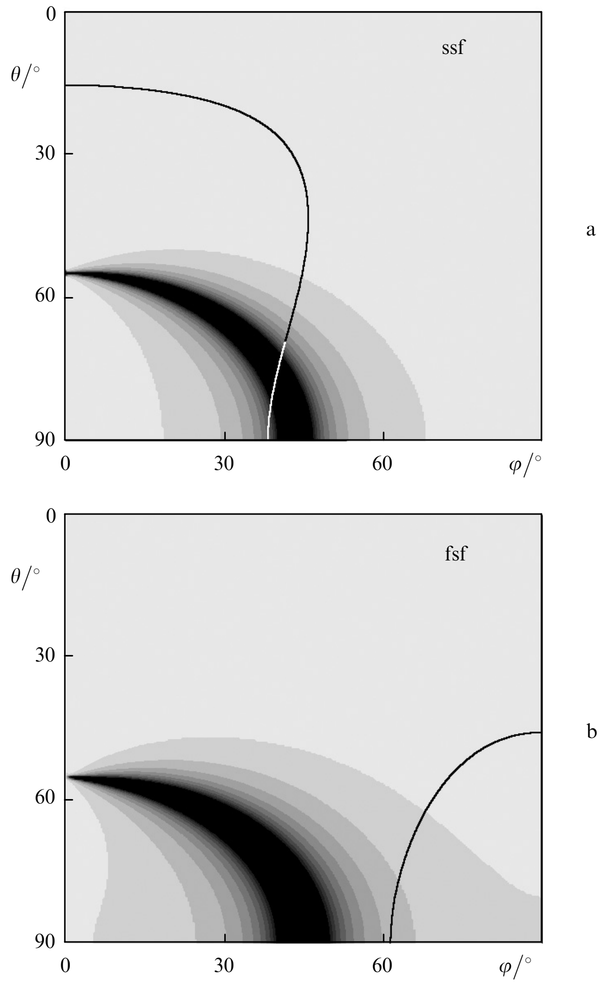

Figure 1. Phase-matching temperature widths for (a) the ssf and (b) fsf interaction and phase-matching directions (thin lines). The black-coloured region indicates a temperature width of > 50 °C.

length different from 1064.2 nm exists for which the point of intersection of the curves lies in the xy plane. For the fsf interaction, the temperature-noncritical THG of the 1064.2-nm radiation is not achieved over the entire range of phase-matching angles (Fig. 1b).

For crystals with large linear expansion coefficients and, primarily, with large anisotropies of these coefficients, one should take into consideration the temperature-strain-induced changes in the crystal as a whole, i.e., the temperature-dependent rotations of its faces. In this case, as follows from (1), the temperature-noncritical phase-matching mode is effected at $\mathrm{d}\Delta\varphi^{(2)}/\mathrm{d}T = \mathrm{d}\varphi_{\mathrm{pm}}/\mathrm{d}T$. However, according to the temperature changes in the refractive indices, the phase matching is temperature-critical, since, in this case, $\mathrm{d}\varphi_{\mathrm{pm}}/\mathrm{d}T < 0$. Since, the inequality $\mathrm{d}\Delta\varphi^{(2)}/\mathrm{d}T > \mathrm{d}\varphi_{\mathrm{pm}}/\mathrm{d}T$ is valid for the LBO crystal in the xy plane, a phase-matching direction can be found for which $\mathrm{d}\Delta\varphi^{(2)}/\mathrm{d}T = \mathrm{d}\varphi_{\mathrm{pm}}/\mathrm{d}T$; i.e., a completely temperature-noncritical phase-matching mode is achieved with all thermal processes taken into consideration (temperature-induced changes in the refractive indices and thermal-strain-induced changes in the crystal). No comprehensive data on the temperature derivatives of the refractive indices $\partial n_i/\partial T$ for various frequencies over the entire LBO-transparency range have been obtained by now (these values for

$\lambda = 632.8$ nm are presented in Ref. [10]), and the available dispersive dependences dn_i/dT have a limited accuracy (e.g., [2, 6]). The absence of data on the second derivatives d^2n_i/dT^2 does not allow the exact temperature width to be calculated for the noncritical mode. For this reason, the quantitative results obtained should be regarded as preliminary.

The linear expansion coefficients and temperature derivatives of the refractive indices are temperature functions. Therefore, temperature changes are accompanied by a change in the relationship between $d\Delta\varphi^{(j)}/dT$ and $d\varphi_{pm}/dT$. Figure 2 shows the temperature–angular dependence of the phase-matching temperature width for the ssf-type THG in the LBO crystal in the xy plane with the two above-considered thermal mechanisms taken into consideration. A nonlinear temperature dependence of the medium parameters leads to a situation in which, when the crystal temperature falls (in particular, to negative temperatures), the width of the phase-matching temperature curve ΔT_{pm} slightly increases. It is infinitely wide at the centre of the wide black band; i.e., the phase matching is temperature-noncritical ($d\Delta k/dT = 0$).

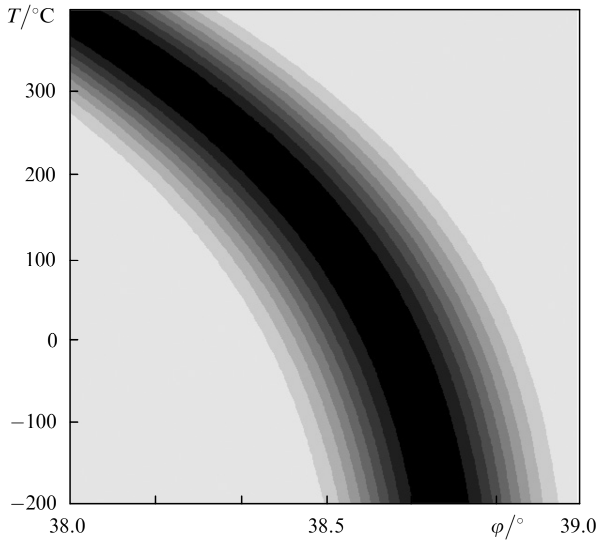

Figure 2. Temperature–angular dependence of the phase-matching temperature width for the THG in an LBO crystal.

This type of dependence of ΔT_{pm} on the crystal temperature was obtained in Ref. [11], in which the temperature-stable THG was achieved for Nd^{3+} : YLF laser radiation (1047 nm + 523.5 nm) in an LBO crystal (no data on the methods of mounting the crystal and on its length are reported). The phase-matching temperature width in Ref. [11] increased from 12 to 67 °C cm, when the crystal temperature dropped from 60 °C to zero.

The above results were verified experimentally for the THG in a 14.1-mm-long LBO crystal grown at the Department of Physics of Moscow State University. The crystal was cut at angles $\varphi = 39°$ and $\theta = 75°$ (ssf interaction). Measurements of the THG of the 1064.2-nm radiation were performed under conditions corresponding to the fixed-field approximation (at a radiation energy of 10 mJ and an average power of 12 mW). The temperature setting accuracy was no worse than 0.1 °C, and the rate of its change was < 300 °C h^{-1}. Figure 3 shows the experimental dependence

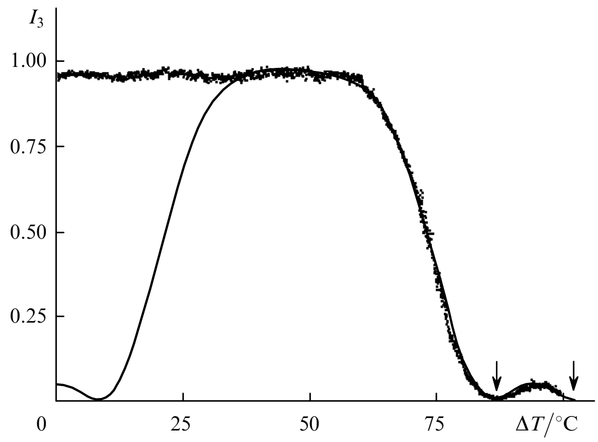

Figure 3. Experimental (dots) and theoretical (the solid curve) phase-matching temperature curves for the THG in an LBO crystal.

of the relative intensity of the third harmonic I_3 on the excess ΔT of the crystal temperature relative to room temperature, which is in fact the phase-matching temperature curve. Its character shows that temperature-noncritical phase matching is achieved; the temperature-curve width (FWHM) is 73 °C. The total width obviously exceeds 73 °C, since part of this curve lies in the region of negative temperatures (the possibility of cooling the crystal during measurements was absent). Thus, from the results obtained, it follows that a temperature-noncritical frequency conversion can be attained not only by cooling the crystal, as was done in Ref. [10], but also at higher temperatures by selecting an optimal cut and a method for fastening the crystal.

It follows from Fig. 3 that the distance between the first and second zeros of the phase-matching curve (shown with arrows) is 15 °C. This means that the peak of the phase-matching curve in Fig. 3 must correspond to the temperature change $\Delta T = 48$ °C under the assumption that this curve has the form of $\operatorname{sinc} x^2$ (the distances of the first and second zeros of this curve from the peak must be 36.3 °C and 51.3 °C, respectively). The phase-matching curve $\operatorname{sinc} x^2$ corresponding to the main contribution of the second temperature derivative of the difference of the refractive indices is shown with the solid curve in Fig. 3. The right branch of the calculated curve is in complete agreement with the measured results. This means that the first temperature derivative of the difference of the refractive indices is zero and the main contribution is due to the second derivative. However, the conversion efficiency remains virtually constant in a temperature range of 0–50 °C. Since the linear expansion coefficients are weak temperature functions, it can be pointed out that the data in Fig. 3 at 0–50 °C evidently correspond to the case where the difference of the temperature derivatives of the refractive indices is equal to zero not only for the first- and second-order derivatives, but for higher orders as well. In this case, the dependence of the phase-matching temperature width on the crystal length is not radical.

The following can be noted in conclusion. The THG process in an LBO crystal is temperature-critical if only the temperature dependence of the refractive indices is taken into consideration. There exists a direction in which a noncritical interaction is ensured only for the difference of the refractive indices. However, in this direction, the contribution of thermal-strain-induced crystal changes, which lead to a detuning relative to the phase-matching direction, is nonzero. The total insensitivity of the frequency-conversion process to the temperature can be ensured by a compensating effect of thermal-strain-induced rotations at an appropriate choice of the method for mounting the crystal. For the THG in a 14.1-mm-long LBO crystal for Nd^{3+}:YAG laser radiation, we have obtained a phase-matching temperature width of at least 73 °C. In crystals with a small contribution of thermal-strain-induced changes, the temperature-noncritical phase matching is achieved with a temperature-noncritical difference of the temperature derivatives of the refractive indices of the interacting waves ($\mathrm{d}\Delta n_i/\mathrm{d}T = 0$).

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