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Integral criterion for selecting nonlinear crystals for frequency conversion

S.G. Grechin

***Abstract*. An integral criterion, which takes into account all parameters determining the conversion efficiency, is offered for selecting nonlinear crystals for frequency conversion. The angular phase-matching width is shown to be related to the beam walk-off angle.**

Keywords: *frequency conversion of laser radiation, phase-matching width, nonlinear crystals.*

Criteria making it possible to determine potentials of crystals for second- and higher-harmonic generation of both continuous-wave and pulsed laser radiation have been formulated for more than a forty-year period of the development of methods for nonlinear optical frequency conversion [1 – 5]. These criteria include a number of parameters defining the conversion efficiency of a nonlinear crystal. To generate the second harmonic in the phase-matching direction (determined by angles θ and φ) in the fixed field approximation, the conversion efficiency has the form

$$\eta_{\text{p}} = 240\pi^3 \frac{d_{\text{eff}}^2}{n^3} \frac{L_{\text{cr}}^2}{\lambda_{10}^2} I_{10}, \qquad (1)$$

where d_{eff} is the effective nonlinearity coefficient; n is the refractive index of the crystal; L_{cr} is the crystal length; λ_{10} and I_{10} are the wavelength and intensity of incident radiation, respectively.

One of widely used parameters of a crystal [figure of merit (FOM)] is the effective nonlinearity coefficient $\text{FOM}_1 = d_{\text{eff}}$. A more accurate parameter is $\text{FOM}_2 = d_{\text{eff}}^2/n^3$, which takes into account the change in the field strength and phase velocity in a crystal with respect to the environment because n^3 for different crystals can differ by several times.

In addition to the FOM_2 parameter characterising nonlinear properties, the phase-matching width governed by optical properties of a crystal is used. Depending on a particular problem, it can be the spectral phase-matching width $2\Delta\lambda_{\text{phm}}$, angular phase-matching width ($2\Delta\theta_{\text{phm}}$ for single-axis crystals, $2\Delta\theta_{\text{phm}}$ or $2\Delta\varphi_{\text{phm}}$ for two-axis crystals) or temperature phase-matching width $2\Delta T_{\text{phm}}$. Factor 2 means full phase-matching width. The angular phase-matching width is usually defined outside the crystal to make it easier to compare it with the laser beam divergence. Parameter $2\Delta\lambda_{\text{phm}}$ is relevant to the frequency conversion of ultrashort laser pulses, broadband radiation or the output of tunable lasers. The least of the two parameters $2\Delta\theta_{\text{phm}}$ or $2\Delta\varphi_{\text{phm}}$ is used in frequency conversion of a multimode or focused laser radiation. Parameter $2\Delta T_{\text{phm}}$ is used in problems of frequency conversion when the crystal temperature needs to be changed considerably.

The relation between the effective nonlinearity coefficient and phase-matching width is different for different crystals, which makes it rather difficult to choose a suitable crystal. However, it is possible to employ one integral parameter taking various properties of crystals into account. Consider, for example, the case when the angular phase-matching width is most important.

The phase-matching width is determined as a linear parameter per crystal unit length. For a crystal of length L_{cr} the angular phase-matching width is

$$2\Delta\theta_{\text{cr}} = 2\Delta\theta_{\text{phm}}/L_{\text{cr}}. \qquad (2)$$

When crystals are compared, the condition that the width of the spatial emission spectrum is equal or does not exceed the angular phase-matching width is used to find the length of the crystals. Using relations (1) and (2), it is possible to determine the integral parameter

$$\text{FOM}_{3\theta} = \frac{d_{\text{eff}}^2(2\Delta\theta_{\text{phm}})^2}{n^3}. \qquad (3)$$

This parameter characterises both nonlinear and phase-matching properties of a crystal. Similarly, it is possible to define parameter $\text{FOM}_{3\lambda}$ allowing for the spectral phase-matching width: $\text{FOM}_{3\lambda} = d_{\text{eff}}^2(2\Delta\lambda_{\text{phm}})^2/n^3$, and FOM_{3T} allowing for the temperature phase-matching width: $\text{FOM}_{3T} = d_{\text{eff}}^2(2\Delta T_{\text{phm}})^2/n^3$. The expression for FOM_{3T} takes into account the total change in the crystal temperature, for example, in the case when the environmental temperature changes. When the limitations are caused by the thermal self-action for which the crystal temperature is defined by the average power of laser radiation and the effective absorption coefficient of the crystal [6], the

S.G. Grechin Scientific Research Institute of Radio-electronics and Laser Technology, N.E. Bauman State Technical University,
2-ya Baumanskaya ul. 5, 105005 Moscow, Russia;
e-mail: gera@bmstu.ru

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expression for the complex parameter FOM_{3T} takes the form:

$$FOM_{3T} = \frac{d_{\mathrm{eff}}^2 (2\Delta T_{\mathrm{phm}})^2}{n^3} \left(\frac{\lambda_T}{\alpha_{\mathrm{eff}}} \right)^2, \quad (4)$$

where λ_T is the thermal conductivity coefficient of the crystal and α_{eff} is the effective absorption coefficient.

Table 1 gives the parameters of some widely-used nonlinear crystals for second-harmonic generation at pump wavelengths 1.0642 and 0.5321 µm. In particular, it follows from the table that apart from $LiNbO_3$ and KTP crystals, $LiIO_3$ and BBO crystals have best FOM_1 and FOM_2 at 1.0642 µm; however, the latter two have worse $FOM_{3\theta}$ than LBO and CLBO crystals. At 0.5321 µm a BBO crystal demonstrates best FOM_1 and FOM_2, whereas DKDP, ADP and CLBO crystals have best $FOM_{3\theta}$. A BBO crystal is inferior to all other crystals because of the smaller angular phase-matching width. At 1.0642 µm the ADP crystals enjoy best $FOM_{3\lambda}$ mostly because of large spectral phase-matching width. CLBO, BBO and LBO crystals lag behind.

In frequency conversion the intensity of incident radiation should not exceed the damage threshold I_{lim} of the crystal. The damage threshold is a characteristic of a medium. Because of this fact, parameter $FOM_{4\theta}$ can be derived from (1). Related to the angular phase-matching width, FOM_4 has the form similar to (3):

$$FOM_{4\theta} = \frac{d_{\mathrm{eff}}^2 (2\Delta\theta_{\mathrm{phm}})^2}{n^3} I_{\mathrm{lim}}. \quad (5)$$

The published values of the damage threshold of nonlinear crystals differ significantly. Table 2 gives typical values of $FOM_{4\theta}$ measured in (ang. minute)^2 at different I_{lim} for the pulse duration of 10 ns at $\lambda_{10} = 1.0642$ µm.

Table 2. Figures of merit $FOM_{4\theta}$ for some crystals.

Crystal	Phase-matching type	I_{lim}/GW cm^{-2}	$FOM_{4\theta}$/(ang. minute)^2
ADP	ooe	1.5	2.02
	oee	1.5	14.1
DKDP	ooe	1.0	0.81
	oee	1.0	6.91
$LiNbO_3$	ooe	0.15	113.2
$LiIO_3$	ooe	0.05	0.43
BBO	ooe	3.0	16.1
	oee	3.0	22.5
CLBO	ooe	6.0	8.7
	oee	6.0	104.9
LBO	ssf	2.5	209.1
KTP	sff	0.15	812.73

While parameter $FOM_{3\theta}$ describes the conversion efficiency of radiation with a finite divergence or in the case of angular misalignment, parameter $FOM_{4\theta}$ allows for the limiting characteristics of the crystal. The relationship between $FOM_{3\theta}$ and $FOM_{4\theta}$ is different for different crystals. $LiNbO_3$ and KTP crystals have large $FOM_{3\theta}$ only for low light intensities (to 150 MW cm^{-2}). When the intensity is high, LBO and CLBO crystals take the lead. In the case of frequency conversion of collimated high-intensity beams when the angular phase-matching width does not limit the conversion efficiency and the main limitation is determined by the damage threshold, figure of merit (5) can be determined by neglecting the parameter $2\Delta\theta_{\mathrm{phm}}$.

In designing commercial frequency-conversion lasers, the issue of cost of nonlinear crystals is rather important.

Table 1. Nonlinear and phase-matching properties of crystals.

Crystal	λ_{10}/µm	Phase-matching type	θ	φ	$2\Delta\theta_{\mathrm{phm}}$	$2\Delta\varphi_{\mathrm{phm}}$	$2\Delta\lambda_{\mathrm{phm}}$/nm	$2\Delta T_{\mathrm{phm}}$/°C	FOM_1	FOM_2	$FOM_{3\theta}$	$FOM_{3\lambda}$	FOM_{3T}
ADP	1.0642	ooe	42°14′6″	45°	5′22″	–	10.57	1.54	0.39	0.0468	1.35	41.4	0.11
	1.0642	oee	62°23′	0, 90°	11′32″	–	54.02	1.82	0.47	0.0711	9.43	207.2	0.24
	0.5321	ooe	78°33′46″	45°	5′57″	–	0.14	0.37	0.57	0.0911	3.22	1.7×10^{-3}	0.013
DKDP	1.0642	ooe	36°33′55″	45°	6′24″	–	5.80	26.76	0.25	0.0197	0.81	0.66	14.1
	1.0642	oee	53°38′23″	0, 90°	11′48″	–	4.96	27.62	0.40	0.0496	6.91	1.22	37.8
	0.5321	ooe	84°34′	45°	12′34″	–	0.14	3.3	0.42	0.051	8.09	1×10^{-3}	0.55
$LiNbO_3$	1.0642	ooe	83°3′17″	90°	17′20″	–	0.32	1.25	6.24	2.51	754.1	0.26	3.92
$LiIO_3$	1.0642	ooe	29°59′26″	–	2′10″	–	0.64	11.21	3.55	1.843	8.65	0.76	231.6
	1.0642	oee	44°0′5″	–	3′41″	–	0.68	8.66	≡ 0	≡ 0	≡ 0	≡ 0	≡ 0
BBO	1.0642	ooe	22°52′39″	30°	2′54′	–	2.12	29.86	1.7	0.6375	5.36	2.86	568.4
	1.0642	oee	32°33′	0	4′30″	–	2.16	31.25	1.27	0.370	7.49	1.72	361.4
	0.5321	ooe	47°24′58″	30°	57″	–	0.074	5.42	1.32	0.3710	0.33	2.1×10^{-3}	10.9
	0.5321	oee	82°12′10″	0	7′05″	–	0.086	6.52	0.03	0.0002	0.01	1.5×10^{-6}	0.0084
BiBO	1.0642	ssf	40°44′	0	1′50″	3°28′	0.76	–	2.37	0.932	3.19	0.538	–
	1.0642	sff	53°50′43″	0	3′36″	5°40′	0.776	–	0.53	0.0466	0.604	0.028	–
	1.0642	sff	41°54′53″	47°	4′30″	11′40″	0.871	–	2.11	0.7385	14.95	0.561	–
CLBO	1.0642	ooe	29°11′52″	45°	5′12″	–	3.86	48.68	0.42	0.0537	1.45	0.8	127.3
	1.0642	oee	42°29′28″	0, 90°	8′40″	–	3.85	49.83	0.86	0.2311	17.49	3.42	573.8
	0.5321	ooe	61°26′52″	45°	2′27″	–	0.13	5.32	0.76	0.1699	1.02	3×10^{-3}	4.81
LBO	1.0642	ssf	90°	11°36′	4°33′	22′25″	3.8	10.33	0.83	0.1666	83.62	2.41	13.93
KTP	1.0642	sff	90°	22°30′	4°30′	59′11″	0.58	23.11	3.08	1.546	5418.2	0.52	825.7

Notes. All values of phase-matching widths are given for 1-cm-long crystals. The values of angular phase-matching widths are given outside a crystal. FOM_1 is measured in pm V^{-1}, FOM_2 (ang. minute pm V^{-2})^2, $FOM_{3\theta}$ (nm pm V^{-1})^2 and FOM_{3T} (°C pm V^{-1})^2.

In most cases it is possible to determine the overall cost of a Cs crystal knowing the price of one cubic millimetre (Pr). Varying with crystals, Pr is dependent on the crystal-growth method, the size and optical quality of a particular crystal. In this case the figure of merit FOM_{Pr} (efficiency – cost) can be introduced instead of FOM_3. For example, $FOM_{3\theta}$ (3) can be replaced by

$$FOM_{Pr\,\theta} = \frac{FOM_{3\theta}}{Cs} = \frac{d_{eff}^2 2\Delta\theta_{phm}}{n^3 Pr}. \qquad (6)$$

Unlike (3), expression (6) holds a linear dependence on phase-matching bandwidth, which says in favour of crystals with larger coefficients of the effective nonlinearity. FOM_{Pr} can also be defined via the spectral and temperature phase-matching widths. Note that apart from the cost of the crystal itself, the overall cost of a frequency-conversion system is determined by the necessity to employ temperature control, housing, coating, etc.

Expressions (3)–(6) do not hold the beam diameter, which means that they are defined for a collimated beam. The frequency conversion of focused radiation requires the beam diameter, the beam divergence and the optimal crystal length to be taken into account.

It is necessary to emphasise another point here. In a number of papers dealing with comparison of nonlinear crystals the authors mention, for instance, that a crystal is characterised by a small angular phase-matching width and large walk-off angle. However, both parameters are nothing else than manifestation of the same property of anisotropic crystals – angular dependence of the refractive index (spatial dispersion of the medium). In the particular case of single-axis crystals (or in the principal planes of two-axis crystals) the walk-off angle is

$$\beta = \frac{1}{n_e(\theta)} \frac{dn_e(\theta)}{d\theta}, \qquad (7)$$

where n_e is the refractive index of an extraordinary wave. In the general case, the width of angular phase-matching outside a crystal is

$$2\Delta\theta_{phm} = \lambda_{10} n \left(\frac{d\Delta n}{d\theta}\right)^{-1}. \qquad (8)$$

In type I phase matching, the angular phase-matching width taking (7) into account has the from

$$2\Delta\theta_{phm} = 100 \frac{\lambda_{10}}{2|\beta_{2\omega}|}, \qquad (9)$$

and in type II phase matching, it is

$$2\Delta\theta_{phm} = 100 \frac{\lambda_{10}}{|2\beta_{2\omega} - \beta_{\omega}|}. \qquad (10)$$

Factor 100 in relations (9) and (10) is introduced because the angular phase-matching width is commonly defined for a 1-cm-long crystal. It follows from (9) and (10) that the angular width for the type II phase matching is always greater than that for the type I phase matching.

One can clearly see that the most objective parameter for describing spatial dispersion is the angular phase-matching width, which, in the most general case, takes into account all terms ($d^m n/d\theta^m$) of the expansion of the refractive index and allows it to be compared to the laser beam divergence, rather than the walk-off angle, which is defined only by the first term of the power series and used for diffraction-quality beams. The expression for the 'aperture length' $L_a = d_0/\beta$ for the laser beam diameter d_0 holds true in the case of single-mode beams and type I phase matching. For multimode beams whose figure of merit is M^2, $L_a = d_0/(\beta_{\omega} M^2)$. For type II phase matching, $L_a = d_0/(|2\beta_{\omega} - \beta_{2\omega}|M^2)$.

This paper gives the definition of the integral criterion for comparison of nonlinear crystals (figures of merit) for nonlinear-optical frequency conversion. The characteristic parameters of widely-used crystals are presented. Figures of merit can be determined in a similar way for the problems of generation of sum and difference frequencies. The relationship between the angular phase-matching width and walk-off angle is demonstrated. All computational results have been obtained with the help of the LID-FC software package (Laser Investigator & Designer – Frequency Conversion) [7].

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