Second harmonic generation in periodically poled crystals for two types of interaction

S G Grechin, V G Dmitriev

Abstract. **Second harmonic generation of laser radiation in periodically poled crystals is considered for two types of interaction simultaneously. The possibility of highly efficient frequency conversion of depolarised laser radiation is demonstrated.**

Periodically poled crystals offer new quantitative and qualitative potentialities for solving the problems of nonlinear optical frequency conversion of laser radiation. One of these involves a considerable increase in the conversion efficiency on account of the nonlinear susceptibility tensor coefficients which are not used in homogeneous crystals since the phase matching is forbidden by crystal dispersion. In addition, the generation of various types of interaction or processes (second and third harmonic generation, sum frequency generation, etc.) can be provided for different orders of quasi-phase matching. We showed [1] that if the quasi-phase-matching condition is satisfied during the second harmonic generation (SHG) for two types of interaction (for example, ssf and sff) in biaxial crystals, quasi-phase matching will be automatically provided for the third type of interaction (fff). The same is true for other combinations of various types of interaction: ffs, sfs, sss in biaxial crystals and ooe, oee, eee and eeo, eoo, oeo in uniaxial crystals. The orders of quasi-phase matching are connected through certain relations. For ooe, oee and eee types of interaction, the following relation holds:

$$m_{oee} = \frac{m_{ooe} + m_{eee}}{2}. \tag{1}$$

Consider the peculiarities of the second harmonic generation for various relations between the quasi-phase-matching orders by the example of a negative uniaxial LiNbO$_3$ crystal of the point group 3m. Fig. 1 shows the dependences of the required domain structure period $\Lambda = ml_c$, where l_c is

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the coherence length [2], on the wavelength λ of the fundamental harmonic for three types of interaction (ooe, oee and eee) with different quasi-phase-matching orders m upon the SHG. Calculations were made by using the Sellmeyer equations from Ref. [3]. For the ooe-type interaction at wavelengths 975.6 and 4233.5 nm, the period tends to infinity, which corresponds to the fulfilment of the phase-matching condition for homogeneous crystals. The quasi-phase-matching order is negative ($m_{ooe} = -1$) within the wavelength range from 975.6 to 4233.5 nm and positive outside it ($m_{ooe} = 1$), which is typical for all negative crystals. Similarly, the phase-matching condition is satisfied for homogeneous crystals in the case of the oee interaction at 1471.9 and 2574.0 nm, the quasi-phase-matching order being negative within this wavelength range and positive outside it.
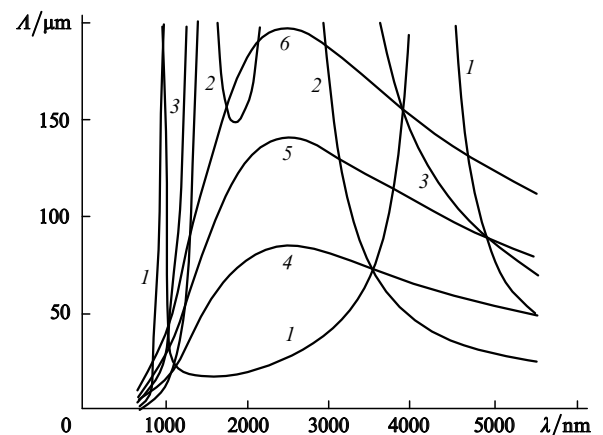

Figure 1. Dependences of the SHG domain structure period on the wavelength of fundamental radiation for the ooe interaction ($|m_{ooe}| = 1$) (*1*), oee ($|m_{oee}| = 1$) (*2*), oee ($|m_{oee}| = 3$) (*3*), eee ($|m_{eee}| = 5$) (*4*), eee ($|m_{eee}| = 7$) (*5*).

Table 1 shows the wavelengths λ of the fundamental harmonic and the quasi-phase-matching orders for which SHG is realised simultaneously for three types of interaction considered by us. One can easily see that the condition (1) is satisfied for all combinations. We confined our analysis to the case when the quasi-phase-matching order is not very high. It is obvious from Eqn (1) that the number of such combinations in the general case is infinite.

The phase-matching orders may have the same sign or opposite signs. Without loss of generality, consider the case when the radiation propagates in the principal xy plane of the crystal. Because the effective nonlinearity of the oee

Table 1.

λ_1/nm	m_{ooe}	m_{eee}	m_{oee}
883.5	+1	+5	+3
1039.91	−1	+7	+3
1139.6	−1	+3	+1
3515.6	−1	+3	+1
3886.27	−1	+7	+3
4886.11	+1	+5	+3

interaction for the point group 3m is identically equal to zero in this case, simultaneous SHG is possible only for the ooe and eee interactions.

Suppose that the radiation propagates along the y axis of the crystal and the plane of polarization of the fundamental radiation is oriented at an angle γ to the x axis in the xy plane. The system of equations in the generally accepted notation (see, for example, Ref. [2]) for the field components of interacting waves (components A_{1x}, A_{1z} of the fundamental harmonic and A_{2x}, A_{2z} of the second harmonic) for all crystal symmetry point groups has the form

$$\frac{\partial A_{1x}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_\omega^o}\left\{d_{11}A_{2x}A_{1x}^*\exp(-i\Delta k_{ooo}z)\right.$$

$$\left. + d_{12}A_{2z}A_{1z}^*\exp(-i\Delta k_{oee}z) + d_{15}[A_{2x}A_{1z}\exp(-i\Delta k_{eoo}z)\right.$$

$$\left. + A_{2z}A_{1x}^*\exp(-i\Delta k_{ooe}z)]\right\},$$

$$\frac{\partial A_{1z}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_\omega^e}\left\{d_{31}A_{2x}A_{1x}^*\exp(-i\Delta k_{eoo}z)\right.$$

$$\left. + d_{33}A_{2z}A_{1z}^*\exp(-i\Delta k_{eee}z) + d_{35}[A_{2x}A_{1z}^*\exp(-i\Delta k_{eoe}z)]\right\},$$

$$\frac{\partial A_{2x}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_{2\omega}^o}\left[d_{11}A_{1x}^2\exp(i\Delta k_{ooo}z)\right.$$

$$\left. + d_{13}A_{1z}^2\exp(i\Delta k_{eeo}z) + 2d_{15}A_{1x}A_{1z}\exp(i\Delta k_{eoo}z)\right],$$

$$\frac{\partial A_{2z}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_{2\omega}^e}\left[d_{31}A_{1x}^2\exp(i\Delta k_{ooe}z)\right.$$

$$\left. + d_{33}A_{1z}^2\exp(i\Delta k_{eee}z) + 2d_{35}A_{1x}A_{1z}\exp(i\Delta k_{eoe}z)\right]. \tag{2}$$

In the particular case of a LiNbO$_3$ crystal for which d_{15}, d_{31} and d_{33} are nonzero, we have

$$\frac{\partial A_{1x}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_\omega^o}d_{15}A_{2z}A_{1x}^*\exp(-i\Delta k_{ooe}z),$$

$$\frac{\partial A_{1z}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_\omega^e}d_{33}A_{2z}A_{1z}^*\exp(-i\Delta k_{eee}z),$$

$$\frac{\partial A_{2z}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_{2\omega}^e}\left[d_{31}A_{1x}^2\exp(i\Delta k_{ooe}z)\right.$$

$$\left. + d_{33}A_{1z}^2\exp(i\Delta k_{eee}z)\right]. \tag{3}$$

For the total amplitudes of the fields of interacting waves, these equations take the form

$$\frac{\partial A_\omega}{\partial z} = -i\frac{\pi}{\lambda_{10}}A_{2\omega}A_\omega^*\left[\frac{d_{15}}{n_\omega^o}\cos^2\gamma\exp(-i\Delta k_{ooe}z)\right.$$

$$\left. + \frac{d_{33}}{n_\omega^e}\sin^2\gamma\exp(-i\Delta k_{eee}z)\right],$$

$$\frac{\partial A_{2\omega}}{\partial z} = -i\frac{\pi}{\lambda_{10}n_{2\omega}^e}A_\omega^2\left[d_{31}\cos^2\gamma\exp(i\Delta k_{ooe}z)\right.$$

$$\left. + d_{33}\sin^2\gamma\exp(i\Delta k_{eee}z)\right],$$

$$\frac{\partial \gamma}{\partial z} = -i\frac{\pi\sin 2\gamma}{4\lambda_{10}}\frac{A_{2\omega}A_\omega^*}{A_\omega}\left[\frac{d_{33}}{n_\omega^e}\exp(-i\Delta k_{eee}z)\right.$$

$$\left. + \frac{d_{15}}{n_\omega^o}\exp(-i\Delta k_{ooe}z)\right]. \tag{4}$$

Here, $A_{2\omega}$ is the modulus of the second harmonic wave amplitude.

The last equation in (4) describes the change in the slope γ due to a difference in conversion processes for both field components of the fundamental radiation. Using the spectral approach for describing the effective nonlinearity [4], we obtain

$$\frac{\partial A_\omega}{\partial z} = -i\frac{2}{\lambda_{10}}A_{2\omega}A_\omega^*\left[\frac{d_{15}}{n_\omega^o m_{\text{ooe}}}\cos^2\gamma\exp(-i\delta k_{ooe}z)\right.$$

$$\left. + \frac{d_{33}}{n_\omega^e m_{\text{eee}}}\sin^2\gamma\exp(-i\delta k_{eee}z)\right],$$

$$\frac{\partial A_{2\omega}}{\partial z} = -i\frac{2}{\lambda_{10}n_{2\omega}^e}A_\omega^2\left[\frac{d_{31}}{m_{\text{ooe}}}\cos^2\gamma\exp(i\delta k_{ooe}z)\right.$$

$$\left. + \frac{d_{33}}{m_{\text{eee}}}\sin^2\gamma\exp(i\delta k_{eee}z)\right],$$

$$\frac{\partial \gamma}{\partial z} = -i\frac{\sin 2\gamma}{2\lambda_{10}}\frac{A_{2\omega}A_\omega^*}{A_\omega}\left[\frac{d_{33}}{n_\omega^e m_{\text{eee}}}\exp(-i\delta k_{eee}z)\right.$$

$$\left. - \frac{d_{15}}{n_\omega^o m_{\text{ooe}}}\exp(-i\delta k_{ooe}z)\right]. \tag{5}$$

Here, $\delta k_i = \Delta k_i - m_i G$ is the generalised wave detuning for the ith interaction; $G = 2\pi/\Lambda$; Δk_i is the wave detuning compensated by the reciprocal lattice vector of the crystal.

For $\gamma = 0$, generation occurs upon the ooe interaction, while for $\gamma = 90\,°$, it occurs upon the eee interaction. For an exact observation of the quasi-phase matching ($\delta k_{ooe} = \delta k_{eee} = 0$), the expression for the effective nonlinearity coefficient d_{eff} has the form

$$d_{\text{eff}} = \frac{d_{15}}{n_\omega^o m_{\text{ooe}}}\cos^2\gamma + \frac{d_{33}}{n_\omega^e m_{\text{eee}}}\sin^2\gamma. \tag{6}$$

It follows from this expression that d_{eff} is maximum when m_{ooe} and m_{eee} have the same sign. If these parameters have opposite signs, $d_{\text{eff}} \to 0$ if $\gamma \to \gamma_{\text{cr}}$, and γ_{cr} is defined by the expression

$$\tan^2 \gamma_{cr} = -\frac{d_{31} m_{eee}}{d_{33} m_{ooe}}. \tag{7}$$

Fig. 2 shows the angular dependences of the conversion efficiency η upon SHG in a $LiNbO_3$ crystal for the ooe, eee, and both types of interaction for identical signs of the phase-matching order ($m_{ooe} = 1$, $m_{eee} = 5$). Independent generation for each type of the interaction was provided by a coordinated variation in the radiation wavelength and the domain structure period at which the generation for the other type of interaction was absent.
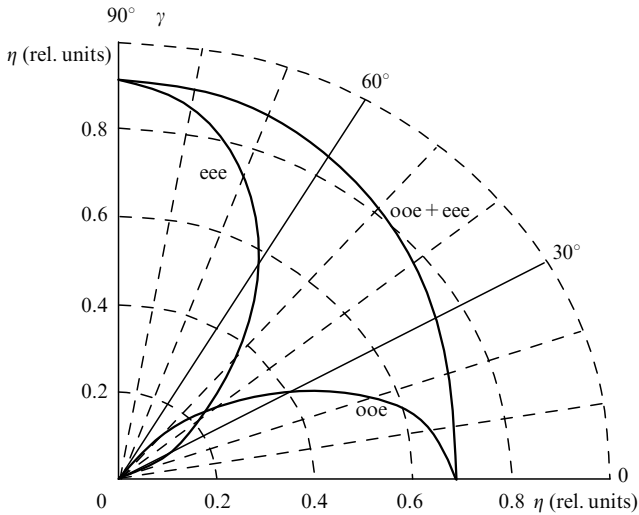

Figure 2. Angle diagram of the dependence of the conversion efficiency η on the angle γ for $m_{ooe} = 1$ and $m_{eee} = 5$.

Because the ratios d_{31}/m_{ooe} and d_{33}/m_{eee} are close in the case under consideration, the total conversion efficiency weakly depends on the angle γ. This means that the fundamental radiation with an arbitrary orientation of the polarisation plane is transformed into linearly polarised radiation of the second harmonic, i.e., an effective SHG can be achieved for the depolarised radiation.

Analogous dependences for SHG for the opposite signs of phase-matching orders ($m_{ooe} = -1$, $m_{eee} = 7$) are shown in Fig. 3. The conversion efficiency is equal to zero for $\gamma = 39.6\,^\circ$. According to the variation in the ratio m_{eee}/m_{ooe}, the relation between the conversion efficiencies at $\gamma = 0$ and $90\,^\circ$ also changes compared to the case shown in Fig. 2.

One of the important questions concerns the stability of the conversion process relative to the generalised wave detuning. In this case, the expression for the coupling coefficient in (5) can be represented in the form

$$d_{eff} = \left\{ \frac{d_{31} m_{eee}}{d_{33} m_{ooe}} \cos^2 \gamma \exp[i(\delta k_{ooe} - \delta k_{eee})z] + \sin^2 \gamma \right\}$$

$$\times \frac{d_{33}}{m_{eee}} \exp(i\delta k_{eee} z). \tag{8}$$

Upon a variation of the wavelength of the fundamental radiation, temperature or the angle of orientation of the crystal, the generalised wave detuning for different types of interaction changes with different rates. It follows from (8) that the conversion process will be characterised by two
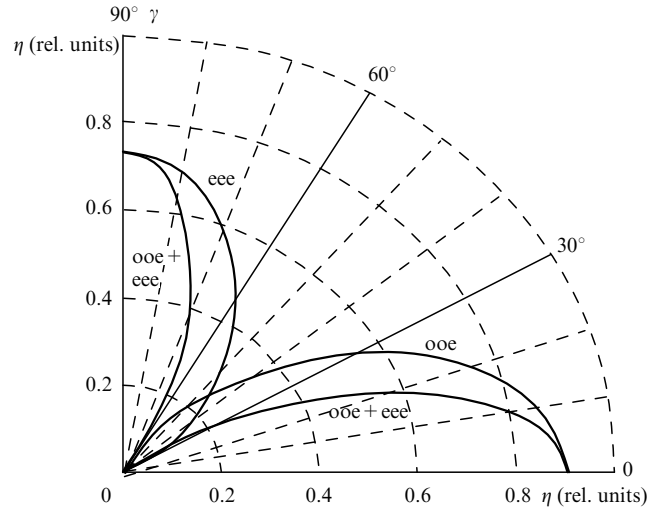
typical spatial beat frequencies associated with the wave detuning (δk_{eee}) and a periodic variation in the nonlinear coupling coefficient ($\delta k_{ooe} - \delta k_{eee}$).

Fig. 4 shows the dependences of the conversion efficiency on the crystal length for an exact quasi-phase matching and in the presence of a generalised wave detuning for $m_{ooe} = 1$ and $m_{eee} = 5$ for $\gamma = 45\,^\circ$.
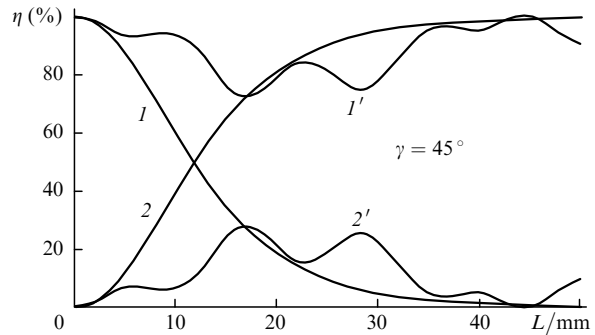

Figure 3. Angle diagram of the dependence of the conversion efficiency η on the angle γ for $m_{ooe} = -1$ and $m_{eee} = 7$.

Figure 4. Conversion of radiation in a periodically poled crystal for exact quasi-phase matching (curves $1, 2$) and in the case of wave detuning (curves $1', 2'$) for fundamental radiation (curves $1, 1'$) and for second-harmonic radiation (curves $2, 2'$).

For $m_{ooe} = -1$ and $m_{eee} = 7$, this detuning leads to the nonzero conversion efficiency at $\gamma = 39.6\,^\circ$. Fig. 5 shows the dependences of the conversion efficiency on the crystal temperature for the two cases considered above. The curves for $\gamma = 0$ and $90\,^\circ$ correspond to SHG for the same type of interaction and differ in the temperature widths because the differences in the temperature derivatives of refractive indices are different. For $\gamma = 45\,^\circ$, the phase-matching curve displays beats defined by the difference in the generalised wave detunings, and does not tend asymptotically to zero.

Consider the variation in the orientation of the polarisation plane of the fundamental radiation during the conversion. If the condition of quasi-phase matching is satisfied, the equation for the angle γ of the polarisation plane has the form
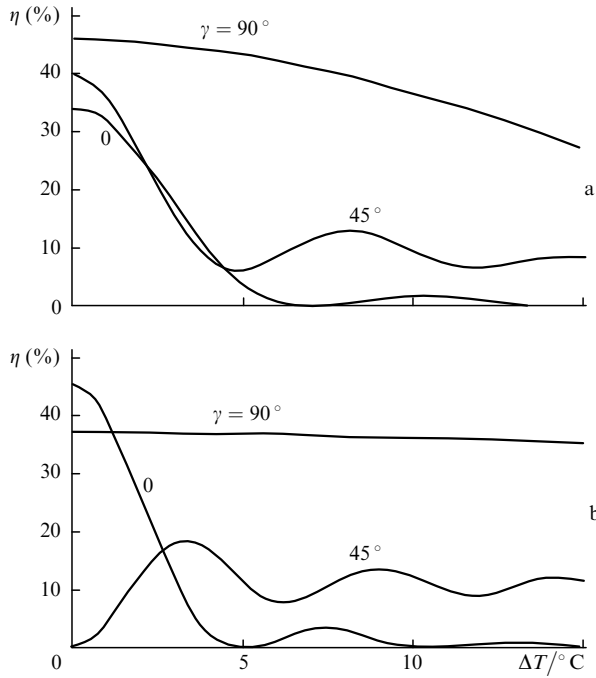
Figure 5. Temperature phase-matching curves for (a) $m_{ooe} = 1$ and $m_{eee} = 5$ and (b) $m_{ooe} = -1$ and $m_{eee} = 7$ for different angles γ.

$$\frac{\partial \gamma}{\partial z} = -i\frac{\sin 2\gamma}{2\lambda_{10}} A_{2\omega}\left(\frac{d_{33}}{n_\omega^e m_{eee}} - \frac{d_{15}}{n_\omega^o m_{ooe}}\right). \quad (9)$$

The fundamental radiation remains linearly polarised during its propagation through the crystal only in two cases, for $\gamma = 0$ and $90°$. The fundamental radiation will be depolarised in all the remaining cases, as well as in the second type of conversion in homogeneous crystals. However, the ratio of the semiaxes of the polarisation ellipse will vary in the problem considered here, and the ellipse will rotate during conversion. The angle of rotation of the polarisation ellipse is decribed by Eqn (9). The equation for the ratio of semiaxes has the form

$$\frac{\partial(A_{1z}/A_{1x})}{\partial z} = \frac{\tan\gamma}{2\lambda_{10}} A_{2\omega}\left(\frac{d_{33}}{n_\omega^e m_{eee}} - \frac{d_{15}}{n_\omega^o m_{ooe}}\right). \quad (10)$$

We have considered above the propagation of radiation in the principal plane xy of the crystal. These results can be easily generalised to the case of an arbitrary direction of propagation, which can be the subject of independent investigations. Here, we note only that for an arbitrary direction of propagation, the effective nonlinearity can be optimised not only over the angle γ and the magnitude and sign of the quasi-phase-matching order, but also over angles θ and φ, which corresponds to multiparametric optimisation of d_{eff}. For example, it can be expected that the maximum value of d_{eff} for a LiNbO$_3$ crystal will correspond to the interval $-\pi/2 < \theta < 0$ of angles, i.e., the conversion efficiency will depend on the octant in which the interaction takes place.

Thus, the analysis of the main features of SHG in periodically poled crystals for several types of interaction simultaneously showed that among the entire set of combinations of the quasi-phase-matching orders for which a simultaneous SHG is possible for several types of inter-

action, the efficient SHG of the depolarised fundamental radiation is possible only for combinations having the same signs.

The calculations were performed by using the reference-computational packet of applied programs LID-SHG (Laser Investigator and Designer-Second Harmonic Generation) located at the site http://www.bmstu.ru/~lid.

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