Periodically poled nonlinear crystals for parametric light generation

E.A. Anfimova,[1,2] P.P. Geiko,[2] and S.M. Shandarov[1]

[1]*Tomsk State University of Control Systems and Radioelectronics*
[2]*Institute of Monitoring of Climatic and Ecological Systems, Siberian Branch of the Russian Academy of Sciences, Tomsk*

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Periodically poled nonlinear crystals (PPNC crystals) with quasi-phase-matching enable efficient generation of a broadband tunable infrared radiation. It is the key property of the PPNC crystals that any constraints on the polarization of interacting waves are removed. Besides, quasi-phase-matching is realizable along any direction with respect to crystal optical axes, which allows maximizing efficient nonlinearity and fitting the domain period to an optimal value. The calculated results are presented on the realization of quasi-phase-matching wave interaction in GaAs, $LiNbO_3$, $KNbO_3$, $CsTiOAsO_4$, $LiTaO_3$, and other crystals as well as periods of nonlinear susceptibility modulation.

Introduction

Use of nonlinear crystals with an induced periodic structure in equipment for spectroscopic remote sensing of the atmosphere allows an essential increase of its functional capabilities. Thus, periodically poled nonlinear crystals (PPNC crystals) with quasi-phase-matching allow broadband radiation to be efficiently generated in the spectral ranges with characteristic absorption spectra of pollutant molecules.[1] In recent years, such PPNC crystals as $LiNbO_3$, $KNbO_3$, $CsTiOAsO_4$, $LiTaO_3$, etc., have been commonly used to generate frequency-tunable radiation in the 3 to 5 μm atmospheric window for solving problems in spectroscopy and remote sensing.[2]

In 1962, N. Blombergen et al., suggested to use the periodic modulation of quadratic susceptibility of crystals to compensate for mismatch between wave vectors of interacting waves. First, the phase synchronism was achieved by use of a stack of plates made of a nonlinear optical material, optical axes of which reverse their directions from plate to plate.[3] Thickness of each plate should be equal to the coherence length at which the amplitude of generated radiation does not decrease. Since the phase matching occurs stepwise, such interaction is called quasi-phase-matching. Wave detuning between interacting waves is compensated for by the vector of reverse nonlinear grid generated by sign modulation of the nonlinear susceptibility. To realize the quasi-phase-matching condition, the length of each domain is to be equal to an even number of coherence lengths.[4]

Later on, this idea was essentially supplemented and developed that resulted in development of up-to-date production technique for high-efficiency crystals with regular and irregular domain structures. Such structures can be produced by means of, for example, repetitive introduction of impurities during crystal growth or by supplying voltage to the periodically located electrodes as well as by the methods of growth.[5]

Physical principles

First of all, note, that PPNC crystals are uniform by linear optical properties in contrast to photon crystals, linear and nonlinear properties of which alter in space.

The scheme of a PPNC crystal with the "head-to-tail" domain orientation is shown in Fig. 1. The polarization of pumping and generated radiation is similar here and is directed along the z-axis.

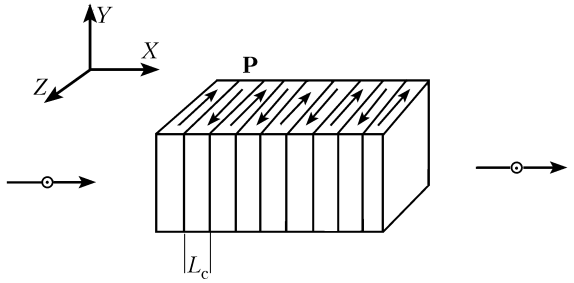

Fig. 1. Nonlinear PPNC crystal: **P** is the polarization vector; all beams are polarized along the Z-axis and propagate along the X-axis.

The principle of realizing the quasi-phase-matching is best understandable by the example of second harmonic generation. In the presence of phase detuning $\Delta k_2 = k_2 - 2k_1$ (k_j is the wave number of pumping radiation and its second harmonic) the complex amplitude of the second harmonic in the preset field approximation varies according to the law[6]

$$\frac{dA_2}{dx} = -i\sigma' A_{10}^2 \exp(i\Delta k_2 x), \qquad (1)$$

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where σ' is the coefficient of nonlinear wave coupling; A_{10} is the pump wave complex amplitude. Integrating Eq. (1), we obtain

$$A_2(x) = i\sigma' A_{10}^2 \frac{\sin(\Delta k_2 x/2)}{\Delta k_2 x/2} \exp(-i\Delta k_2 x/2). \quad (2)$$

The intensity of the second harmonic $I^2 \sim |A_2|^2$ is maximum at the length $x = L_c$; here $L_c = \pi/|\Delta k_2|$ is the coherence length. In this case, the phase shift due to the wave detuning equals π. Place the second nonlinear crystal of the length L_c after the first one, which produces the same wave detuning Δk_2 in the direction of interacting waves propagation. Then the second harmonic amplitude at the crystal output equals to

$$A_2(x) = -(\sigma' - \sigma'')A_{10}^2 \frac{\sin(\Delta k_2 x/2)}{\Delta k_2 x/2}. \quad (3)$$

Here σ'' is the nonlinear coupling coefficient for the second crystal. The amplitude of the second harmonic is evidently maximum if $\sigma' = -\sigma''$. Thus, phase detuning occurring in the first crystal can be compensated for by changing the nonlinearity sign. Amplitude and intensity of the second harmonic can be written in the following form:

$$A_2 = -\frac{2}{\pi}\sigma'(2L_c)A_{10}^2; \quad I_2 = \left[\frac{2}{\pi}\sigma'(2L_c)\right]^2 I_{10}^2. \quad (4)$$

That is, the second harmonic intensity at quasi-phase-matching behaves in the same way as in uniform media with the effective nonlinear coefficient $\sigma_{eff} = 2\sigma'/\pi$. The phase of the second harmonic varies from layer to layer in contrast to uniform media where this phase takes the steady-state value immediately. This argumentation is correct for the second harmonic generation and quasi-phase-matching of the first order, but can easily be generalized for both quasi-phase-matching of higher orders (when $\Delta k_2 = m\pi/L_c$, m is the order of quasi-phase-matching) and other types of three-frequency interactions.

Calculated results

The principal advantage of the PPNC crystals is the absence of any constraints on the polarization of interacting waves, i.e., all interaction types are possible in them, in particular, six types for uniaxial crystals (*oo-o*, *oo-e*, *oe-e*, *eo-e*, *ee-o*, *ee-e*) and six types for biaxial ones (*ss-s*, *sf-s*, *sf-f*, *ss-f*, *ff-f*, *ff-s*). Besides, quasi-phase-matching is realizable in them along any direction with respect to optical axes of crystals, which allows maximizing the efficient nonlinearity and fitting the domain period to an optimal value if necessary.

However it is most important, from the practical point of view, that it becomes possible to use the highest nonlinearity coefficient at quasi-phase-matching by choosing proper polarization of the interacting waves.

For example, the *ee-e*-interaction (all waves are extraordinary) is used in the periodically poled lithium niobate crystal; the component of nonlinear susceptibility d_{33}, exceeding other crystal components by an order of magnitude, is responsible for the interaction. That means that using domain structures the coefficient d_{33} can be phase matched and the conversion efficiency increases by $(2d_{33}/\pi d_{15})^2$ times.

Use of domain structures has also allowed to remove most of the constraints on the frequency and temperature ranges of conversion. The Table compares characteristics of nonlinear properties of the most common monodomain and polydomain crystals.

Characteristics of some nonlinear crystals

Crystal	Transmission band, μm	Uniform nonlinear materials		Periodical nonlinear materials	
		d_{ij}, d_{eff}, pm/V	d^2/n^3, $(pm/V)^2$	d_{ij}, d_{eff}, pm/V	d_{eff}^2/n^3, $(pm/V)^2$
$LiNbO_3$	0.33–5.5	$d_{31} = -4.64$ $d_{22} = 2.46$ $d_{oo\text{-}e} = d_{31}\sin\theta - d_{22}\cos\theta\sin\varphi$	1.95	$d_{33} = 30$; 16	26
$KTiOPO_4$	0.35–4.5	$d_{24} = 3.7$ $d_{oe\text{-}o} = d_{24}\sin\theta$ (XZ plane)	2.6	$d_{33} = 16$; 9.8	18.2
$LiTaO_3$	0.28–5.5	$d_{31} = 3$ $d_{22} = 0.85$ $d_{oo\text{-}e} = d_{31}\sin\theta - d_{22}\cos\theta\sin\varphi$	0.9	$d_{33} = 19$; 11	12.5
$CsTiOAsO_4$	0.35–5.3	$d_{32} = 3.4$ $d_{31} = 2.1$ $d_{oe\text{-}o} = d_{32}\sin\theta$ (XZ plane)	1.7	$d_{33} = 18.1$; 11.5	19.3
$KNbO_3$	0.4–5.6	$d_{31} = -11.9$ $d_{oo\text{-}e} = d_{31}\sin\theta$ (XZ plane)	13.3	$d_{33} = -20.6$; 11	39
$KTiOAsO_4$	0.35–5.3	$d_{32} = 4.2$ $d_{31} = 2.8$ $d_{oe\text{-}o} = d_{32}\sin\theta$ (XZ plane)	3.0	$d_{33} = 16.2$; 9.9	17
$RbTiOAsO_4$	0.35–5.8	$d_{32} = 4.6$ $d_{31} = 1.4$ $d_{oe\text{-}o} = d_{32}\sin\theta$ (XZ plane)	3.6	$d_{33} = 15.9$; 10.1	17.5
$RbTiOPO_4$	0.35–4.5	$d_{32} = 3.8$ $d_{31} = 2.0$ $d_{oe\text{-}o} = d_{32}\sin\theta$ (XZ plane)	2.6	$d_{33} = 15.6$; 9.9	17.7
GaAs	0.9–17	–	–	$d_{36} = 150$; 96	220

As is follows from the Table, polydomain crystals excel the monodomain ones in the nonlinear coefficient proportional to the conversion efficiency.

Solid-state Nd:YAG and $Ti^{3+}:Al_2O_3$ lasers were considered as sources of pump for optical parametric oscillators. Figure 2 shows the calculated results on domain structure periods for 8 crystals.

The structure period is evidently to be from 20 to 45 μm. In principle, it is possible to create structures for the preset wavelengths. Another one advantage of the PPNC crystals is the possibility of providing successive interactions among harmonics, e.g. simultaneous generation of the second and third harmonics and parametric light generation.[7]

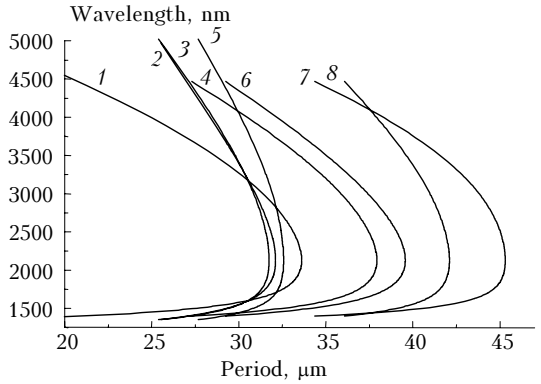

Fig. 2. Lengths of generated signal and idler waves as functions of the domain period in pumping the $CsTiOAsO_4$ (curve *1*), $LiTaO_3$ (*2*), $LiNbO_3$ (*3*), $RbTiOPO_4$ (*4*), $KNbO_3$ (*5*), $KTiOPO_4$ (*6*), $KTiOAsO_4$ (*7*), and $RbTiOAsO_4$ (*8*) crystals by Nd:YAG laser radiation (λ = 1.064 μm).

Smooth frequency tuning is also realizable by choosing proper structure period and varying crystal temperature. The results calculated for this case are shown in Fig. 3.

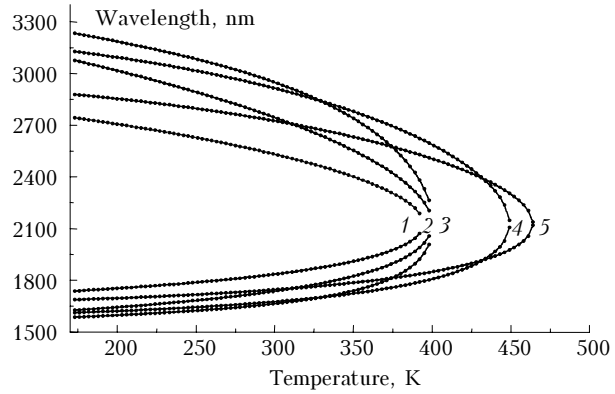

Fig. 3. Lengths of generated signal and idler waves as functions of temperature in pumping crystals by Nd:YAG laser radiation (λ = 1.064 μm): $KTiOPO_4$ crystal with the period of 39 μm (curve *1*); $RbTiOAsO_4$, with the period of 41.5 μm (*2*); $KNbO_3$, with the period of 32 μm (*3*); $LiNbO_3$, with the period of 31 μm (*4*); and $LiTaO_3$, with the period of 31.5 μm (*5*).

The temperature dependences of the refractive indices have been taken from Ref. 8; such dependences are unknown for $RbTiOPO_4$ and $KTiOAsO_4$ crystals. The realistic temperature range from –100 to 200°C has been chosen for the calculations.

Use of smoothly frequency-tunable radiation, e.g. of Ti:Sapphire lasers, is of interest for pumping optical parametric oscillators. As an example, the calculated results for $CsTiOAsO_4$ crystal with different domain periods (from 20 to 25 μm) are shown in Fig. 4. Even at a fixed domain structure period (e.g., 20 μm), radiation within the 1.04–4.45 μm range can be generated by varying wavelength of pumping radiation from 770 to 1000 nm.

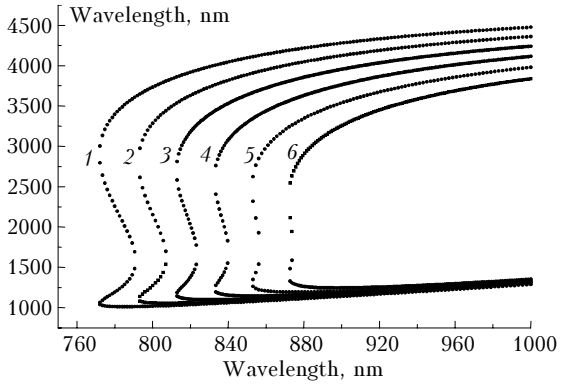

Fig. 4. Lengths of generated signal and idler waves as functions of wavelength of tunable pumping radiation ($Ti^{3+}:Al_2O_3$ laser) in $CsTiOAsO_4$ crystal at the domain structure period of 20 (curve *1*), 21 (*2*), 22 (*3*), 23 (*4*), 24 (*5*), and 25 μm (*6*).

The long-wave IR region 8–12 μm is inaccessible for commonly used PPNC crystals due to strong multiphoton radiation absorption. GaAs is one of the most attractive crystals but it is bad for use as a frequency converter with traditional birefringent synchronism. Its transmission band is 0.9–17 μm, nonlinear optical coefficient d_{14} = 94–150 pm/V exceeds the corresponding coefficient d_{14} = 75 pm/V for $ZnGeP_2$ crystal, which is the most effective birefringent crystal in the middle IR. Large thermal conductivity along with good mechanical properties make GaAs attractive for frequency conversion of high-power laser radiation. We have defined the quasi-phase-matching conditions for wave interaction in GaAs crystals. Lengths of generated signal and idler waves as functions of structure period for the parametric oscillators pumped at the wavelengths of 1.7, 2.1, and 2.8 μm are given in Fig. 5.

If using Nd:YAG laser for pumping and quasi-phase-matching of the first order the period should vary from 15 to 21 μm.

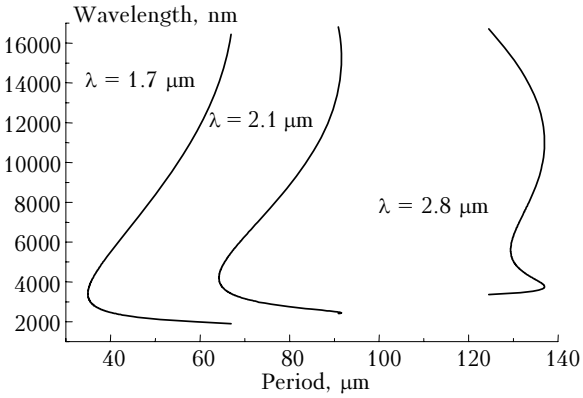

Fig. 5. Wavelength of generated radiation as a function of GaAs domain period at different wavelengths of pump radiation.

The photon efficiencies for conversion of pump radiation to the 3–12 μm region have been calculated for the case of parametric generation (they amount to tens percent). The lasing threshold has been also

estimated (in μJ) when using nanosecond pump pulses. For a crystal of 1 cm length, spectral synchronism widths equal to 2–3 cm^{-1} while temperature synchronism widths are ≈ 2.5 K.

Conclusion

Thus, PPNC crystals are considered to be quite promising for producing frequency-tunable radiation; interaction types unrealizable in crystals, using traditional phase-matching methods, can be realized in these crystals. The state-of-the-art of technology development allows easy production of periodical structures with a period of tens of μm.

Acknowledgments

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