ENGINEERING OF OPTICAL SUPERLATTICE IN LiNbO3:MgO CRYSTAL FOR SIMULTANEOUS SHG OF TWO WAVELENGTHS

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1. Introduction

A new class of materials, called metamaterials with beforehand designed properties, are currently of paramount importance. The most striking examples of metamaterials are photonic crystals and left-handed materials [1, 2]. Such materials with designed parameters are obtained on the base of various superstructures and superlattices created artificially in different media. Layered structures, created in nonlinear-optical crystals for realization of various frequency conversion processes, are called optical superlattices (OSL) [3]. The concept of the OSL allows the creation of compact, multifunctional and efficient laser systems which are in great demand for design compact multicolor sources of coherent radiation, in quantum nonlinear optics, optical communications, biomedicine, remote sensing, etc. In ferroelectric non-linear-optical crystals such OSL is possible to get by fabrication of domain structures, arranged according to determined regularity either during crystals growth or by special post-growing technologies. Most works in this area have been done with ferroelectric LiNbO₃ (LN), LiTaO₃ (LT) and KTiOPO₄ (KTP) crystals developed for nonlinear optics [4-6]. They are commercially available in large, homogeneous single-domain crystals and wafers, and can be domain-inverted to give a stable structure with a more or less ideal modulation of the nonlinearity.

In this work, multi-wavelength interactions, that can be implemented in crystals with nonlinear coupling coefficient changes according to an aperiodic law, are studied. A theoretical model is considered based on detailed analysis of coupled equations for amplitudes of interacting waves in nonlinear materials [7]. The model was used to calculate an optimal construction of domain blocks and the regularity of their consecutive sequence in the LN:MgO crystal capable to fulfill simultaneously phase-matching (PM) conditions for two second harmonic generations (SHG) of radiations at the 1064 and 1320 nm. The addition of MgO into the matrix is necessary for strong suppression of the photorefractive (optical-damage) effect inherent to LN crystals [8].

2. Theory

To achieve an efficient frequency conversion for all nonlinear processes it is necessary to fulfill simultaneously energy and momentum conservation laws, providing an effective energy exchange between the interacting waves with ω_1 , ω_2 and ω_3 frequencies and \mathbf{k}_1 , \mathbf{k}_2 and \mathbf{k}_3 wave-vectors:

$$h\omega_1 + h\omega_2 = h\omega_3$$
$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3.$$

The second law in nonlinear optics is known as phase-matching (PM) condition for interacting waves in a nonlinear medium. PM condition can be provided either by the birefringence of nonlinear material or by use the quasi-phase matching (QPM) technique, i.e. with involvement a reciprocal lattice vectors of material structures [9]. The QPM technique can be realized on periodic, quasi periodic or aperiodic (APOSL) OSL, created artificially in nonlinear-optical materials (Fig.1).

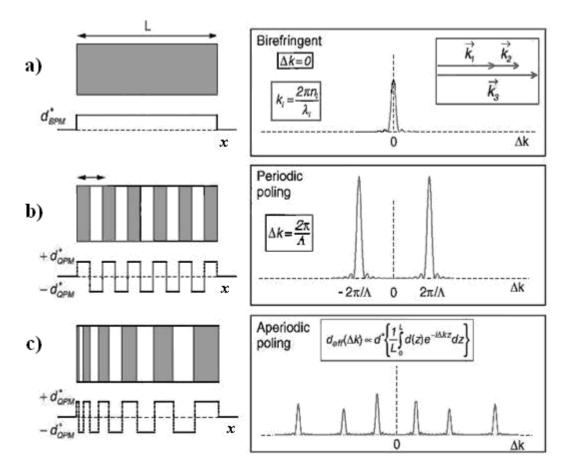


Fig.1. Spatial distribution of nonlinear coefficients (left) and their Fourier transforms (right) for the birefringent (a), periodically poled (b) and aperiodically poled (c) ferroelectric crystals.

The main condition for all types of QPM methods is a creation of the OSL provided reciprocal wave vectors, compensating phase mismatch between interacting waves. To determine a

reciprocal lattice vector of a structure, the Fourier transformation (FT) technique is used. For onedimensional POSL with the Λ period, the FT gives

$$F_{POS}(x) = \sum_{m} f_{m} \exp(iG_{m}x).$$
(1)

Conversion coefficients are determined by the following relations:

$$f_m = (2/m\pi)\sin(m\pi/2); \quad \Delta k = k_{2\omega} - 2k_\omega - G_m; \quad G_m = 2\pi m/\Lambda .$$
⁽²⁾

Here k_{ω} and $k_{2\omega}$ are wave numbers of interacting waves, G_m are reciprocal lattice vectors of the OSL. Since the POSL provides the set of reciprocals with integral values of the *m* factor then it, obviously, can be used only for the QPM of fundamental wave harmonics. In order to realize multi-frequency QPM, the QPOSL and APOSL were developed. A QPOSL presents itself an array of two periodic OSL, the periods' ratio of which is irrational number. The lattice structures are formed from two structured A and B blocks consisting of positive and negative domains with specific thicknesses and ranked in concrete sequences. The set of optical frequencies, which can be simultaneously PM, is richer than with the POSL [10]. However, for any frequencies it is impossible to satisfy simultaneously PM conditions.

The main advantage of the APOSL is a possibility to design structures allowing the implementation of QPM for any frequencies with preassigned relative amplitudes of coefficients of the FT, as well as the use of the largest nonlinear optical coefficient of the material. By the FT the coefficients efficiencies of corresponding frequency conversion nonlinear processes are defined [9-14]. Thus, the key problem is to design the appropriate APOSL on the base of LN:MgO, by which two reciprocals $G_{1,2}$ should be provided to phase-match the SHG processes of radiations at 1064 and 1320 nm at the same time.

Hereinafter we will follow the theoretical approach of numerical calculations developed in the works [11-14]. When a single laser beam with a frequency ω is incident onto an APOSL, then in order to use the largest nonlinear coefficient (d_{33} for the LN), it is necessary to choose the walls of each domain to be parallel to the *yz* plane, the propagation and the polarization directions of incident light to be along the *x* and *z* axes, respectively (Fig.2).

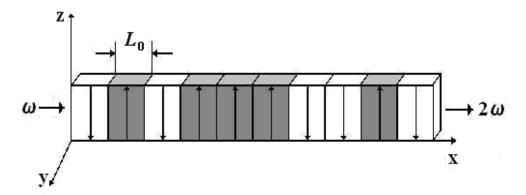


Fig.2. Geometry to use the largest nonlinear coefficient d_{33} of the LN crystal.

Adopting the small-signal approximation, when the depletion loss of fundamental wave power is neglected, the SH conversion efficiency η_{SHG} can be expressed as

$$\eta_{SHG} = \frac{8\pi^2 |d_{33}|^2 I_{\omega} L^2}{c\epsilon_0 \lambda^2 n_{\omega}^2 n_{2\omega}} \left| \frac{1}{L} \int_0^L g(x) \exp[i(k_{2\omega} - 2k_{\omega})x] dx \right|^2,$$
(3)

where *c* is the speed of light in vacuum, ε_0 the permittivity of vacuum, n_{ω} ($n_{2\omega}$) the refractive index of fundamental (SH) wave, *L* the sample total length, k_{ω} ($k_{2\omega}$) the wave number of the fundamental (SH) wave, and g(x) only takes binary values of 1 or -1. To solve the inverse solution problem and design the required OSL, it is useful to introduce a reduced effective nonlinear coefficient for SHG:

$$d_{eff}(\lambda_m) = \frac{1}{L} \left| \int_0^L g(x) \exp\left[i2\pi \frac{x}{l_c(\lambda_m)} \right] dx \right|.$$
(4)

Here *m* numerates different wavelengths and $l_c(\lambda) = (n_{2\omega}-n_{\omega})\lambda/2$ is the coherence length for SHG. If the thickness of the unit block is Δx , then the number of blocks in sample is $N = L/\Delta x$. The position of each block is located at $x_q = q\Delta x$, for q = 0, 1, 2, ..., N-1. The integral equation (4) can be expressed as

$$d_{eff}(\lambda_m) = \frac{1}{N} \left| \operatorname{sinc}\left[\frac{\Delta x}{l_c(\lambda_m)}\right] \sum_{q=0}^{N-1} g(x_q) \exp\left\{ i \left[\frac{2\pi(q+0.5)\Delta x}{l_c(\lambda_m)}\right] \right\} \right|,$$
(5)

where $\sin c(x) = \sin(\pi x)/(\pi x)$. Obviously, the optimal construction of the APOSL for the SHG can be ascribed as a search for the maximum of $d_{\text{eff}}(\lambda_m)$ with respect to $\beta(q\Delta x)$. The maximum value condition corresponds to the appearance of the perfectly constructive interference. Such an optimization nonlinear problem can be solved with a mathematical optimization procedure over output intensities of the primary wavelengths with additional condition to have desired rations of intensities of output waves (for example, approximately equal). Numerical calculations were carried out by use of the Monte Carlo method's modification, i.e. by simulated annealing Metropolis algorithm (SAMA) [15] and then the required arrangement of the domain orientations of the blocks in the sample under study was determined completely.

The objective function in the SA method is chosen as

$$E = \sum_{m=1}^{2} \left[\left| d^0 - d_{eff}(\lambda_m) \right| \right] + \left[\max\left\{ d_{eff}(\lambda_m) \right\} - \min\left\{ d_{eff}(\lambda_m) \right\} \right], \tag{6}$$

where the symbol max{...}(min{...}) manifests to take the maximum (minimum) value among all the quantities including into {...}. Numerical calculations by the iterative algorithm for a LN:MgO sample with $L \approx 2$ mm and N = 600 were performed to find out the construction of the APOSL, needed to achieve two-wave SHG for selected wavelengths of incident light ($\lambda_1 = 1.064 \mu m$, $\lambda_2 = 1.320 \mu m$) with equal intensities. Refractive indices of the LN:MgO crystal were calculated according to the Sellmeier formula [16]. A gray scale diagram of a part of the constructed APOSL is presented in Fig.4. The black (white) strip represents the positive (negative) domain. In Fig.5 the calculated reduced effective nonlinear coefficients in the spectral range under study are displayed. One can see two strong peaks with almost identical peak value at the predesignated 532 nm and 660 nm wavelengths.

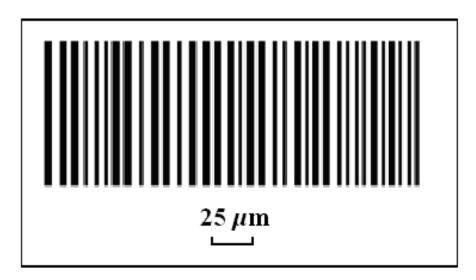


Fig.4. A part of the calculated APOSL.

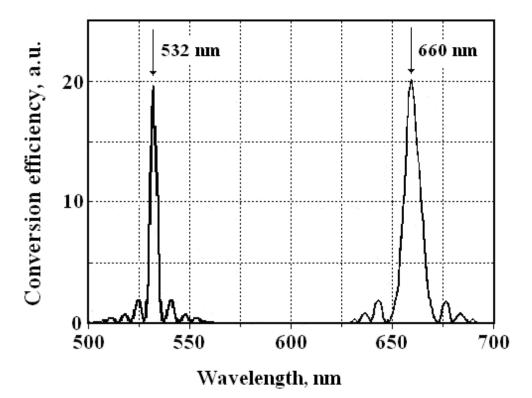


Fig.5. Fourier spectra of the calculated APOSL.

3. Conclusions

In summary, the presented study has allowed numerical definition of the APOSL structure designed for efficient and simultaneous implementation of two SHG nonlinear processes in the

LN:MgO crystals. To find out the desired construction of the APOSL inverse source mathematical optimization problem was solved by use of numerical calculations via the SAMA algorithm. The derived results will be employed for an aperiodic domain structure fabrication in post-growth treatments by applying external electrical high-voltage pulsed technique [17, 18].

It is obvious that various theoretical approximations and applied models will caused some deviations of calculated values from true ones. In order to obtain a more precise definition of required law and structure of domain blocks further adjustments are planned by use of electrooptical changes of refractive indexes of constituent parts of the domain blocks [19, 20]. However, the wide scatter of data in values of domain thicknesses and the absence of analytic expression for spatial distribution function of domains, obviously, will require the development of the special experimental technology to reproduce exactly such structures on LN:MgO crystals in practice.

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