ISSN 0030-400X, Optics and Spectroscopy, 2009, Vol. 107, No. 3, pp. 359–362. © Pleiades Publishing, Ltd., 2009.
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SOLID-STATE LASERS AND NONLINEAR FREQUENCY CONVERSION

Nonlinear Optical Processes in Domain Structures of Strontium Tetraborate

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Received January 22, 2009

Abstract—The random quasi-phase-matching in irregular domain structures of strontium tetraborate has been investigated for nonlinear optical generation at 266 nm. The factor of increasing second-harmonic generation efficiency due to random quasi-phase-matching is determined. The spectral dependences of the second-harmonic generation are calculated. It is shown that its efficiency can be optimized at a fixed pump wavelength by rotating the domain structure.

PACS numbers: 42.70.Mp, 42.25.Dd, 42.65.Ky, 77.80.Dj

DOI: 10.1134/S0030400X09090070

The known ways of effective generation imply phase matching, i.e., that the interacting waves have equal phase velocities in the nonlinear medium. Nevertheless, this condition is not always satisfied in view of certain limitations. Efficient conversion can also be implemented using quasi-phase-matching [1] in regular domain structures, i.e., alternating regions with oppositely oriented static polarization vectors. However, it is fairly difficult to prepare strictly periodic structures. The deviation of real domain structures from perfect structures (providing quasi-phase-matching) was considered in [2]. The analysis showed that various deviations lead to phase mismatch, which deteriorates the conversion efficiency. It was predicted in [3] that, in contrast to the quadratic increase in structures responsible for exact quasi-phase-matching, the conversion efficiency in strongly disordered domain structures should increase linearly with the sample thickness. This prediction was experimentally confirmed by the example of a polycrystalline ZnSe sample, in which the domain thicknesses somewhat differed from the mean value (equal to the coherence length) [4]. The theoretical consideration of highly disordered structures, where the size of individual domains deviated, according to the Gaussian distribution, by 1, 10, and 32% from the mean (generally equal to the coherence length) leads to similar conclusions [5]. The type of matching implemented in these irregular domain structures was referred to as random quasi-phase-matching.

The recently found domain structures in strontium tetraborate (SBO) crystals [6] are characterized by a higher degree of disorder than the structures considered in [4, 5]. In this paper, we report the results of an experimental study of collinear second-harmonic generation using irregular SBO domain structures and their calculated spectral characteristics.

Strontium tetraborate is a promising nonlinear optical material due to the unique combination of properties: its transparency range is limited by 125 nm in the UV region [7], and it has a fairly high nonlinear susceptibility [7, 8]. In addition, SBO is characterized by a high optical breakdown threshold [9], due to which it can be used for nonlinear optical conversion of femtosecond pulses. Despite a fairly low birefringence, femtosecond SHG was implemented in SBO [7] in the absence of phase matching. Ferroelectric crystals are used to form regular domain structures. To date, there are no data indicative of ferroelectric properties of SBO. Nevertheless, irregular domain structures are formed in SBO crystals during growth, which can be used for nonlinear optical conversion [8]. The sample under study contained domain structures highly ordered along the crystallographic *b* and *c* axes but strongly disordered along the crystallographic *a* axis. A fragment of the domain structure, revealed by chemical etching, is shown in Fig. 1.

Random quasi-phase-matching is observed upon propagation of pump radiation through an irregular SBO domain structure along the crystallographic *a* axis. Using the approach developed in [4, 5], one can calculate the total amplitude of the field induced by individual domains from the expression

$$E_{2\omega} = \sum_{n=1}^{N}\left[\frac{2\omega^2\chi_n^{(2)}}{k_{2\omega}(\theta)\Delta k(\theta)}E_\omega^2\{\exp[i\Delta k(\theta)d_n]-1\}\right.$$
$$\left.\times\exp\left(i\Delta k(\theta)\sum_{r=n+1}^{N}d_n\right)\right], \quad (1)$$

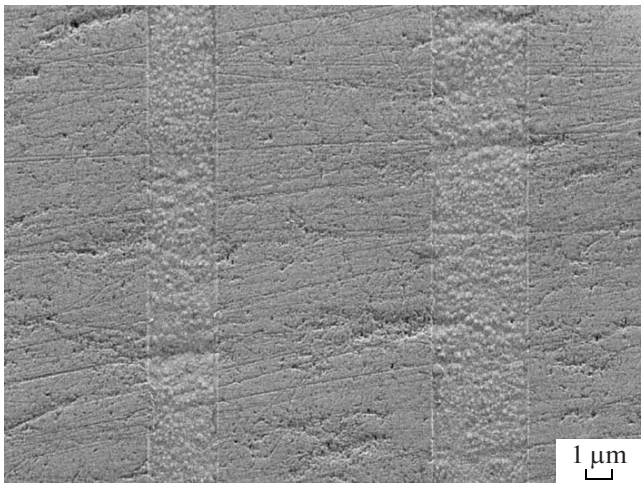

Fig. 1. Fragment of SBO domain structure after chemical etching (JEOL JSM 7001 electron microscope).

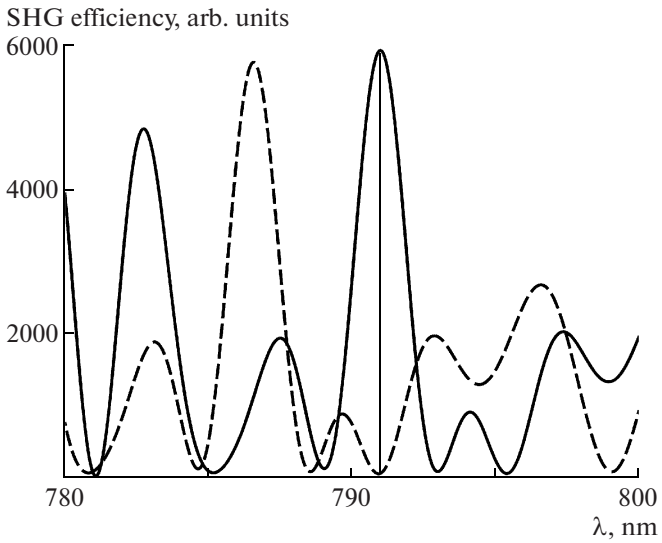

Fig. 2. SHG efficiency near the maximum of the Ti:sapphire laser tuning curve for different angles of incidence: (dotted line) radiation propagates along the crystallographic *a* axis ($\theta = 0°$) and (solid line) the crystal is rotated by the angle $\theta = 9.6°$.

where d_n is the thickness of an individual domain; $\chi_n^{(2)} = (-1)^n|\chi^{(2)}|$ is its nonlinear second-order susceptibility; θ is the internal angle of rotation of the domain structure; and $\Delta k(\theta)$ is the phase mismatch wave vector, which takes into account the anisotropy of refractive indices.

When radiation with a fixed wavelength is converted, the interference of the contributions of all domains to the induced field may be completely destructive (Fig. 2) at the corresponding wavelength (791 nm). In this case, by rotating the crystal, one can modify the spectral dependence so as to provide a maximum at the required wavelength.

Theoretically, this conversion can be described using formula (1) modified as follows:

$$E_{2\omega} = \sum_{n=1}^{N}\left[\frac{2\omega^2\chi_n^{(2)}}{k_{2\omega}(\theta)\Delta k(\theta)}E_\omega^2\left\{\exp\left[i\Delta k(\theta)\frac{d_n}{\cos\theta}\right]-1\right\}\right.$$
$$\left.\times\exp\left[i\Delta k(\theta)\sum_{r=n+1}^{N}\frac{d_r}{\cos\theta}\right]\right]\sqrt{F(\theta_{ext})}, \quad (2)$$

where θ is the internal angle and $\sqrt{F(\theta_{ext})}$ is the envelope factor, which is calculated for SBO from the formula

$$F(\theta_{ext}) = \frac{\left|P_2^{NL}\right|^2 w^{(2\omega)}(w^{(\omega)}+\cos\theta_{ext})}{(w^{(2\omega)}-w^{(\omega)})^2(w^{(2\omega)}+w^{(\omega)})(w^{(2\omega)}+\cos\theta_{ext})^3}, \quad (3)$$

where θ_{ext} is the external angle, $P_2^{NL} = \cos^2\theta_{ext}/(w^{(\omega)} + \cos\theta_{ext})^2$, $w^{(2\omega)} = (n_2^{(2\omega)2} - \sin^2\theta_{ext})^{1/2}$, and $w^{(\omega)} = (n_2^{(\omega)2} - \sin^2\theta_{ext})^{1/2}$.

The envelope factor (3) takes into account the angular dependence of the refractive indices in an anisotropic crystal, the angular change in nonlinear coefficients, and the angular dependence of the Fresnel reflection coefficients. In general, it is fairly difficult to calculate this factor. However, we used the fact that (according to our data) the domain walls are fairly thin (Fig. 1), and the domain structure can be considered homogeneous for the linear optical coefficients. Thus, we used the formula for the envelope factor derived for a homogeneous crystal of *mm*2 symmetry [10].

Figure 2 shows the results of calculating the SHG radiation at a wavelength near the maximum of the Ti:sapphire laser tuning curve. It can be seen in Fig. 2 that, at a corresponding choice of the angle of rotation, the interference of the domain contributions can be changed from completely destructive to more constructive. At small angles of rotation, as a rule, the spectral dependence is shifted; however, a simultaneous shift and change in the shape of the spectral curve may generally occur [11]. Thus, it is of interest to study the angular dependences of random quasi-phase-matching.

This dependence was experimentally studied using 532-nm pump radiation to implement SHG at a wavelength of 266 nm. The theoretical and experimental angular dependences of the signal at doubled fre-

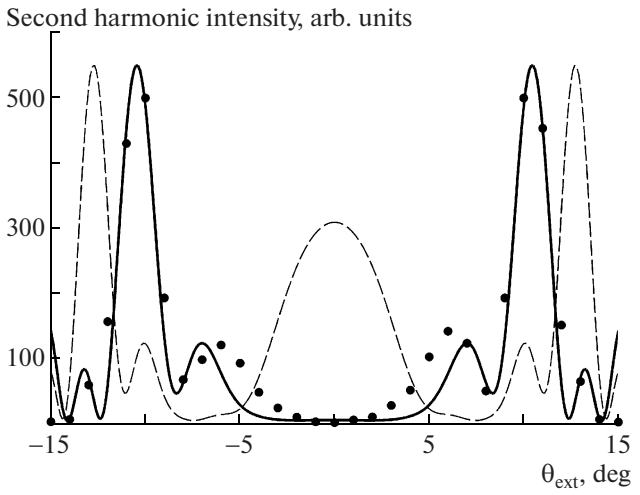

Fig. 3. Angular SHG dependence under random quasi-phase-matching in an irregular SBO domain structure: (circles) experimental data, (dotted line) theoretical curve for the calculated Δk, and (solid line) theoretical curve for $\Delta k = 1.0035\Delta k_{calc}$.

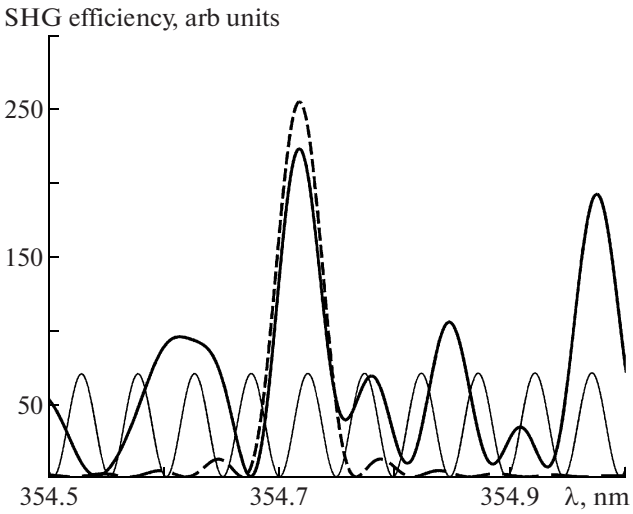

Fig. 4. Spectral dependence of the SHG efficiency at wavelengths near 354.7 nm: (thin solid line) a single-domain sample (multiplied by 500), (bold solid line) a sample with an irregular domain structure rotated by 3.1°, and (dotted line) a regular domain structure (divided by 7000).

quency upon crystal rotation around the pump polarization axis are shown in Fig. 3. These dependences are in good agreement with the results of theoretical calculations from formula (2) at angles of rotation of −15° to 15°. Initially, to plot the theoretical curve, we used the wave mismatch calculated with refractive indices derived from the Sellmeier formula; the coefficients were taken from [9]. In this case, the calculated and experimental dependences were in poor agreement. The calculated curve is highly sensitive to variations in Δk. In this context, we used the least-squares method to compare the experimental and calculated data. The wave mismatch was taken to be a variable because the error in its determination is controlled by the error in approximating the refractive indices. The best agreement was observed at $\Delta k = 1.0035\Delta k_{calc}$. To determine the increase in the SHG efficiency due to random quasi-phase-matching, a 432-μm-thick, single-domain sample was taken as the reference. The SHG efficiency at random quasi-phase-matching in an irregular domain structure corresponding to an angle of rotation of 10° increased by a factor of 501 with respect to the SHG efficiency in a single-domain sample under normal incidence of radiation. The experimental increase in the SHG efficiency nearly coincides with the calculated one (500).

SHG radiation in the near-UV range (particularly at a wavelength of 354.7 nm) is most interesting for practice because it falls in the strong absorption band for most known nonlinear crystals, except for SBO. Figure 4 shows the calculated SHG efficiency near this wavelength. This result indicates that, to obtain the maximum SHG efficiency in the samples under study at this wavelength, the domain structure must be rotated by 3.1°. In this case, the SHG efficiency under random quasi-phase-matching increases by a factor of 1600 in comparison with mismatched conditions. For comparison, Fig. 4 shows the SHG efficiency for an ideal regular domain structure of the same thickness as the irregular structure in the samples studied. This efficiency exceeds that for our samples by a factor of 8000; however, as was mentioned above, the technology of such regular SBO structures is still absent. Hence, according to the aforesaid, it should be developed. Note that the spectral width of the random quasi-phase-matching peak at a wavelength near 355 nm in the domain structure under consideration is about 100 GHz. The quasi-phase-matching peak has the same width in the ideal regular domain structure. This means that, here, the limitations on the spectral width for a regular domain structure and for a randomized sample studied are nearly identical.

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research, project nos. 07-02-00497; grants NSh-1011.2008.2 and NSh-3818.2008.3 of the President of the Russian Federation in Support of Leading Scientific Schools, grant RNP.2.1.1.3455; DPS project 2.5.2; and the Siberian Branch of the Russian Academy of Sciences project nos. 5 and 27.1, and projects "Scientific and educational specialists of innovational Russia."

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Translated by Yu. Sin'kov