Controllable narrow band and broadband second-harmonic generation by tailored quasiphase matching with domain gratings

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We demonstrate second-harmonic generation for various discrete input wavelengths by a controllable quasiphase matching method. This is done by forming ferroelectric domain gratings in $Sr_xBa_{1-x}Nb_2O_6$ crystals. The method allows flexible preparation of tailored quasiphase matching for prespecified wavelengths over prespecified bandwidths. Conversion efficiencies of 5% were achieved.

Quasiphase matching[1–4] provides an important way to obtain efficient optical second-harmonic generation (SHG). However, the common methods for its implementation comprise an irreversible forming of periodic structure in the nonlinear media; hence its operation is restricted to a specific input wavelength. Recently[5,6] we presented a new method that relaxes this limitation by introducing a controllable spread spectrum quasiphase matching method that gives broadband second-harmonic generation. This was accomplished by forming domain gratings in $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN). We have also used this method to fix images and holograms in the crystal. The fixing is based on a photoscreening mechanism that imprints an intensity light pattern (obtained, for example, by two interfering beams) in the ionic structure of the crystal, by forming ferroelectric domains. A spread spectrum of the domain gratings' period produces the broadband SHG capability, demonstrated for input wavelengths between 750 and 1064 nm.

In this letter[7] we report on the observation of a narrowband SHG with well-defined ferroelectric domain grating periodicities. Here, the random broadening of the domain grating structure of our former work is eliminated. We also demonstrate the capability of this method to form tailored quasiphase matching for several wavelengths, or for prespecified wavelength ranges. The quasiphase matching is accomplished by inducing the domain structures with interfering light beams. The method is flexible and controllable and is easily performed in real time.

The experimental setup for the fixing process is identical to that described in Ref. 5. Two mutually coherent beams of an argon ion laser (514.5 nm) with equal intensities (10 mW) illuminated a poled $Sr_{0.75}Ba_{0.25}Nb_2O_6$ (SBN) crystal, with dimensions of $3\times3\times4$ mm^3 and c axis along the 4 mm side. The crystal was nominally undoped. Both light beams were extraordinarily polarized and were focused on the crystal via a cylindrical lens ($f=10$ cm) to form a light layer, which was perpendicular to the crystal c axis and the applied field direction. This makes the screening process effective. The interfering beams formed a grating with a wave vector which was also perpendicular to the c axis. However, in order to create ordered domains and prevent the formation of the pseudorandom microdomains that were a part of the process in our former work, we used an undoped crystal and illuminated a wider layer of the crystal (1 mm instead of 30 μm in Ref. 5). (Then, the pseudorandom microdomains, previously formed due to nonuniform filamentations of the voltage drop in the illuminated light layer, are less likely to be induced.) Most of the other parts of the fixing process were similar to our former work, described in Ref. 5: After poling the crystal with 2.5 kV for 5 min we illuminated the crystal and applied an external voltage along the c axis which had an opposite polarity to that used in the crystal poling. This voltage was gradually increased from 500 to 1100 V over 10 s. Then, the writing beams were blocked and the polarity of the applied voltage was inverted. The voltage was increased from 400 to 700 V in 5 s. Unlike the fixing process for holograms which was described in Ref. 5 we proceeded to apply high voltage on the crystal for about 2 min. This was done in order to allow the domains to spread over the crystal. The size of that spreading can be controlled by changing the duration of this last step. Then a confined volume can be obtained with applied electric field (to induce a refractive index difference) and may be used for waveguiding the beams and increasing significantly the conversion efficiency of the SHG. The fixing could be erased by illuminating the crystal uniformly while applying high voltages (3 kV).

We were able to control the period of the grating, λ_g, by changing the angle between the writing beams. The needed period for quasiphase matching must be equal to the "coherence length" of the SHG:

$$\lambda_g=\frac{\lambda}{2[n(2\omega)-n(\omega)]},$$

where λ is the wavelength of the fundamental wave and $n(\omega)$ and $n(2\omega)$ are the refractive indices of the fundamental and the second-harmonic waves, respectively. We have calculated the dependence of the refractive index on the wavelength by the Sellmeier dispersion relation;[8,9]

$$n_e^2(\lambda)=1+\frac{S_i\lambda_i^2}{1-(\lambda_i^2/\lambda^2)},$$

with the parameters of Ref. 7 for $Sr_{0.75}Ba_{0.25}Nb_2O_6$: $S_i\lambda_i^2=3.861$, $\lambda_i=0.2$ μm. λ is the wavelength and $n_e(\lambda)$ is the index of refraction for the extraordinary polarization, which we have had for both, input and second-harmonic waves, in order to use the nonlinear coefficient d_{33} which is twice as large as d_{31}.[5,8] However, we could fit the domain

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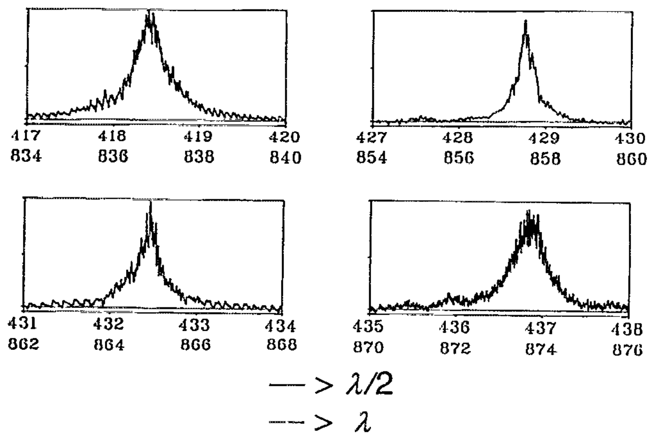

FIG. 1. Power of SHG (in arbitrary units) as a function of the input and the second-harmonic wavelengths (in nm) for several fixed grating periods. A null output was obtained after each erasure of fixed gratings.

periodicity for the d_{31} coefficient,[5] where the input and second-harmonic waves have different polarizations, ordinary and extraordinary, respectively.

The SHG was measured using a pulsed Ti-sapphire laser. The pulses had a duration of 10 ns, a repetition of 10 Hz, and an average energy of 200–400 μJ. The light from the Ti-sapphire laser propagated along a principle axis (in order to avoid the problem of beam walkoff) which was perpendicular to the c axis and parallel to the grating wave vector. In the experiment, the second-harmonic output intensity was measured as the input laser wavelength was scanned by a computer. Figure 1 shows the intensity of the SHG as a function of the fundamental and second-harmonic wavelengths for four different fixed gratings. The peak powers were at input wavelengths, λ=836.8, 857.5, 864.8, and 873.7 nm, corresponding to second-harmonic wavelengths, 418.4, 428.75, 432.4, and 436.85 nm, respectively. The bandwidth of about 0.3 nm is close to the theoretically expected value,[2] showing that we had good periodic gratings along most of the crystal length. Erasure of the gratings after each step was easily done, as in Ref. 5. Then, the SHG signals were eliminated and we had a null output. Figure 2 shows the theoretical

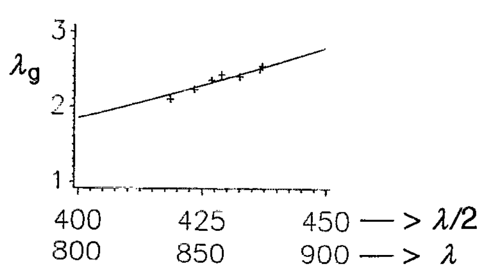

FIG. 2. Theoretical and experimental periods (in μm) of the domain gratings needed to achieve quasiphase matching as a function of the wavelength (in nm) of the input and second-harmonic beams. The crosses show the experimental results. The line is the calculation for the case where both the fundamental and the second-harmonic waves have extraordinary polarization and they propagate along a principle axis which is perpendicular to the c axis.

(according to the expression for λ_g, given above) and the experimental periods of the domain gratings, needed for quasiphase matching, as a function of the wavelengths of the input and second-harmonic beams. The gratings' periods are around 2.0–2.5 μm. The experimental values correspond to the peak power of the second-harmonic beam in each case.

The second-harmonic beam had a confined profile, unlike the angular spread (in the direction perpendicular to the plane which contained the incident wave and the c axis) in the spread spectrum case (broadband SHG) of Ref. 5. The conversion efficiencies reached about 5% for the input beam energies mentioned above (pulses of 200 μJ having a duration of 10 ns), where the beam had quite a distorted profile and a diameter of 1 mm, and the crystal had a length of only 3 mm. However, much higher efficiencies are expected to be obtained after optimizing the input beam and the crystal qualities and by using confined geometries for high light intensities along longer distances.

The multiwavelength capability can be used to obtain quasiphase matching for any prespecified wavelength range, discrete or continuous. It can be exactly tailored to a wavelength range of a tunable laser or to a large linewidth of a

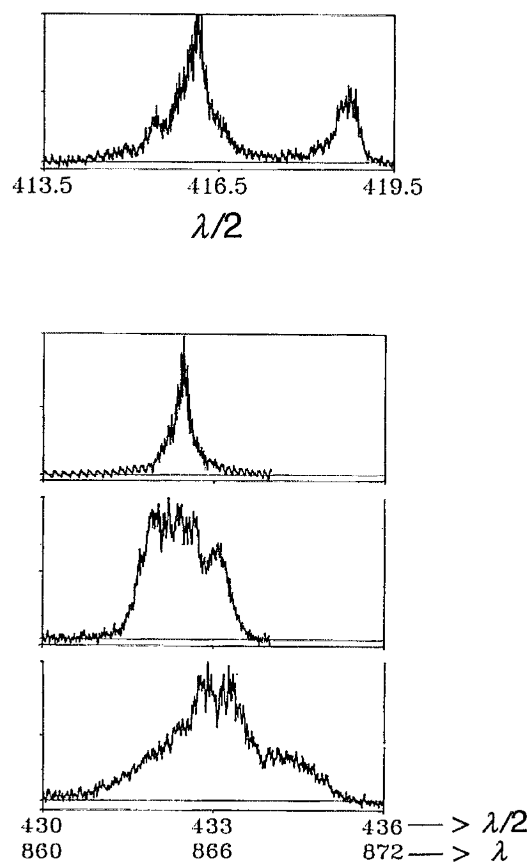

FIG. 3. SHG output as a function of wavelengths (λ and λ/2) with various tailored gratings for quasiphase matching: (a) for two different wavelengths (enabled by two different gratings that simultaneously exist in the crystal); (b) for narrow (as in Fig. 1) and increased bandwidths (obtained when one of the writing beams that induce the gratings in the fixing process was taken to be slightly spherical rather than a plane wave).

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laser diode. The needed grating can be obtained simply by interfering light beams that induce the domain structure. For example, Fig. 3(a) shows the output where two different periodicities were induced in the crystal. The broadening capability of the wavelength acceptance range for SHG is shown in Fig. 3(b). This was obtained by taking one of the gratings "writing" beams to be slightly spherical rather than a plane wave used in the narrowband case. (This technique has some limitations and can produce slight variations in the conversion efficiency, transverse to the beam axis, and hence act to slightly distort the SHG beam.) The bandwidth was increased from 0.2 to 1.6 and 2.3 nm. This capability can make the method attractive for use with lasers which are not very stable or have relatively high linewidths such as laser diodes. It is possible to induce other light patterns (as we have done for images and holograms fixing),[6] that may serve more complicated phase matching needs in the frequency or the spatial domains. We can think of inducing and fixing curved gratings that spatially match nonuniform or complex laser sources.

In conclusion, a demonstration of a controllable narrowband SHG is given. The method allows a flexible tailoring of quasiphase matching to specific wavelengths, prespecified tuning ranges, and frequency widths. This relaxes requirements on specific single wavelength and narrow linewidth operation of SHG, and can be useful for frequency doubling of tunable lasers and laser diodes.

[1] N. Bloembergen, *Nonlinear Optics* (Benjamin, London, 1982).
[2] M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, IEEE J. Quantum Electron. **QE-87**, 2631 (1992).
[3] K. Yamamoto, K. Mizuuchi, and T. Taniuchi, Opt. Lett. **16**, 1156 (1991).
[4] K. Shinozaki, Y. Miyamoto, H. Okayama, K. Kamijoh, and T. Nonaka, Appl. Phys. Lett. **58**, 1934 (1991).
[5] M. Horowitz, A. Becker, and B. Fischer, Appl. Phys. Lett. **62** (1993).
[6] M. Horowitz, A. Becker, and B. Fischer (unpublished).
[7] B. Fischer and M. Horowitz, Opt. Lett. (submitted).
[8] A. M. Prokhorov and Y. S. Kuzminov, *Ferroelectric Crystals for Laser Radiation Control* (Higler IOP, England, 1990).
[9] S. H. Wemple and M. Di Domenico, J. Appl. Phys **40**, 720 (1969).

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