Quasi-phase matching via femtosecond laser induced domain inversion in lithium niobate waveguides

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We demonstrate an all-optical fabrication method of quasi-phase matching structures in lithium niobate (LiNbO₃) waveguides using tightly focused femtosecond near-infrared laser (wavelength 800 nm). In contrast to other all-optical schemes that utilize a periodic lowering of the nonlinear coefficient $\chi^{(2)}$ by material modification, here the illumination of femtosecond pulses directly reverses the sign of $\chi^{(2)}$ through the process of ferroelectric domain inversion. The resulting quasi-phase matching structures, therefore, lead to more efficient nonlinear interactions. As an experimental demonstration we fabricate a structure with the period of 2.74 μm to frequency double 815 nm light. A maximum conversion efficiency of 17.45 % is obtained for a 10 mm long waveguide. © 2016 Optical Society of America

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Quasi-Phase matching (QPM) [1] that employs a spatial modulation of the second-order nonlinear coefficient $\chi^{(2)}$ is an important technique in nonlinear optics. It not only enables efficient frequency conversion, but also makes diverse applications possible including beam and pulse shaping, all-optical processing, entangled photons generation and manipulation. The QPM, i.e. the modulation of nonlinearity $\chi^{(2)}$ can be realized through periodic domain inversion in ferroelectric bulk crystals [2–5], or in waveguide when high intensities are required over a long interaction length [6–8]. For this purpose the electric field poling [9] is commonly used, but it suffers from difficulty of fabricating submicron-sized domains due to the sideways growth effect [10]. Furthermore, the electric field poling cannot be used in the cases of thin films or ferroelectrics deposited onto non-conductive substrates.

A completely different method for ferroelectric domain engi-

neering is all-optical poling, which uses intense laser radiation to directly invert domains [11]. This approach becomes particularly interesting as the light field can be manipulated more accurately with a resolution up to the diffraction limit and hence enables fabrication of fine ferroelectric domains with better defined details. While initially the UV radiations are usually used in all-optical poling, the strong absorption of UV light by ferroelectrics restricts inverted domains into a shallow surface layer (submicrons) [11]. This limits the application of such optically created ferroelectric domain patterns.

Recently, it has been reported the fabrication of QPM structures in ferroelectrics using near-infrared femtosecond laser radiations [12, 13], at which the ferroelectric is transparent. Instead of domain inversion which reverses the sign of the nonlinearity from $+\chi^{(2)}$ to $-\chi^{(2)}$, it is the material modification that is utilized in these schemes to reduce the value of $\chi^{(2)}$ at every coherent length [12, 13]. The problem of this type of QPM structure is that the conversion efficiency is rather low (of the order of a few percents) and the propagation loss due to light scattering is very high (tens of dB/cm) which is much larger than those based on ferroelectric domain inversion. To solve these problems we have recently demonstrated an all-optical poling of ferroelectrics using near-infrared ultrashort pulses [14]. The tight focusing of intense infrared pulses ensures high temperature gradient through multi-photon absorption and, consequently, high strength of the pyroelectric poling field which inverts domains. Using this method we have fabricated 5×7 domain arrays in a congruent LiNbO₃ crystal.

In this letter, we report on the application of our technique to produce a 10 mm long periodic domain pattern in channel waveguide (contains more than 3500 periods) and demonstrate its performance as efficient QPM frequency converter. The second harmonic generation (SHG) is realised with a conversion efficiency of 17.45 %, which is up to 440 times larger than that achieved in the pure waveguide channel without any domain pattern. We use waveguide geometry instead of bulk crystal as the spatial light confinement enables us to maintain high light intensity and, consequently strong nonlinear interaction through the whole sample.

In the experiment the Ti-indiffused LiNbO₃ channel wave-

uide was formed on the -Z surface of a 500 μm -thick congruent crystal. It was fabricated by diffusing a 35 nm thick Ti stripe with a width of 3 μm into the crystal surface, using a diffusion time of 22 hours and a diffusion temperature of 1010°C. The Ti waveguide was designed to be single mode at the non-linear optical pump wavelength of 815 nm, with a refractive index contrast of approximately $\Delta n = 0.001$ and mode depth of $\sim 3 \mu\text{m}$. The loss of the waveguide was measured to be around 0.1 dB/cm for both, fundamental and second harmonic TM modes.

For the fabrication of the inverted domains, the waveguide sample was mounted on a translational stage that can be positioned in three orthogonal directions with a resolution of 100 nm. The infrared light for domain inversion was generated by a femtosecond oscillator (MIRA, Coherent) operating at 800 nm, with a pulse duration of 180 fs, repetition rate of 76 MHz, and pulse energy up to 5 nJ. The light was focused by a 40 $\times$ microscope objective (NA= 0.65) and the diameter of the focus spot on the crystal surface is estimated around 1 μm [Fig. 1 (a)]. For each inverted domain, the focal spot of the laser was translated through the waveguide from -Z towards +Z-surface with an average speed of $v = 10 \mu\text{m/s}$. An automatic shutter was used to block the laser beam when the sample moved to the next region of domain inversion.

Fig. 1 (b) displays a typical image of the obtained two dimensional ferroelectric domain pattern after 5 minutes of etching in hydrofluoric (HF) acid. Prior to etching the sample was annealed at 200 °C for 30 minutes to remove any residue of photorefractive effect from the domain inversion process. The average QPM period is $\Lambda = 2.74 \mu\text{m}$ (along X-axis), aimed at frequency doubling of 815 nm light. In the transverse direction (Y-axis) we realized domain reversion in a period as short as 1.15 μm . We also used the Čerenkov second harmonic microscopy [15, 16] to visualize the 3D domain pattern and confirmed the inverted domains extending as deep as 28 μm [see Fig. 1(c)] below the surface, therefore ensuring a good overlap with the waveguide modes of fundamental and second harmonic.

We showed before [14] that infrared ultrafast laser poling originates from the same underlying physical mechanism as the UV laser direct writing of domains in lithium niobate [17, 18]. It is the presence of thermoelectric (or/and pyroelectric field) in the focal volume of the light that acts as a cause of domain inversion. In the particular case of the infrared poling, the crystal is heated through the process of multi-photon absorption. While the used wavelength 800 nm is too long for band to band two photon absorption (the band gap of LiNbO₃ is $\sim 4 \text{ eV}$), the process could involve two or higher order photon absorption from defect or impurity states within the gap.

The annealed LiNbO₃ waveguide with inscribed domain patterns was used to generate quasi-phase matched second harmonic wave. We used a NA=0.1 microscopic objective to focus the 815 nm laser beam from the femtosecond oscillator (MIRA by Coherent) into the waveguide and collected the emitted second harmonic using a NA=0.2 microscopic objective. The polarization of the fundamental wave is parallel to the Z-axis of the LiNbO₃ sample (TM waveguide mode) to ensure that the largest nonlinear coefficient d_{33} is used in frequency conversion process. Figure 2 shows the far field beam intensity distribution of the fundamental and second harmonic light after passing through the QPM structure. It can be seen that beam shape is well maintained in the frequency doubling process and that in both cases the the fundamental mode of the waveguide is

excited, allowing for a high mode overlap between the fundamental and the second harmonic waves.

Fig. 1. (a) Schematic of direct writing ferroelectric domain patterns in Ti-indiffused LiNbO₃ channel waveguide using femtosecond infrared pulses. (b) The optical microscopic image of the two-dimensional optically poled domain pattern with the period of 2.74 μm in X direction and 1.15 μm in Y direction. Individual inverted domains are visible as small circles. Waveguide boundaries are indicated with dashed lines. (c) Three-dimensional profiles of the inverted domains obtained by the Čerenkov second harmonic microscopy [15, 16].

The temperature tuning was used to optimize the frequency doubling process and it turned out that the maximal harmonic output occurred at 62.5 °C (see Fig. 3). The measured temperature acceptance bandwidth is about 5 °C, which is wider than theoretically predicted by using the Sellmeier equation for LiNbO₃ crystal [19]. It is worth noting that the theoretic result represents the continuous wave case, while the experiment was performed with 150 fs pulses. The spectral width of such ultrashort pulses is around 15 nm. When the temperature of waveguide is tuned away from the optimal value at which the central wavelength is quasi-phase matched, the contributions from the other spectral components to the second harmonic generation grow as they become phase matched. This results in the broadening of the experimental temperature acceptance bandwidth [20–22]. In addition, the group velocity mismatch between the fundamental and second harmonic pulses restricted the effective interaction distance of the second har-

Fig. 2. Output intensity distribution of the fundamental (a) and second harmonic waves (b) in far field. The coordinate system is that of the LiNbO₃ crystal.

Fig. 3. The wavelength tuning response of the second harmonic generation in optically poled LiNbO₃ waveguide. The squares depict experimental results while the narrow curve represents theoretical tuning curve of 10 mm long ideal periodic structure for continuous wave case.

monic generation [21–23], which is about 83 μm in our case. The shorter the effective length is, the wider the acceptance bandwidth becomes [24]. Another major factor responsible for the broadening is the imperfection of the produced domain structures. It is known that in any poling processes the random period errors are unavoidable. Such random deviations from the optimal QPM period will also shorten the effective interaction length of nonlinear processes and broaden the acceptance bandwidth [24].

Fig. 4 shows the output average power and conversion efficiency of the quasi-phase matched SHG process as a function of the input power of fundamental wave. The power of second harmonics follows the square law for low input powers. However, the growth slows down above 85 mW of input power as a consequence of back conversion. A second harmonic power of 15.28 mW was obtained for 87.55 mW of input power. This corresponds to conversion efficiency of 17.45 % and the normalized conversion efficiency of $199.28 \text{ \%W}^{-1}\text{cm}^{-2}$. It should be noted that because our experiments were conducted with short pulses, the frequency conversion process was adversely affected by the group velocity mismatch between the fundamental and second harmonic pulses [21–23]. The group velocity mismatch restricted the effective interaction length of a nonlinear process. The second harmonic wave generated within this length can grow coherently. Beyond this length, the newly generated harmonic wave is essentially incoherent with the previously formed but it still contributes to the total output. Therefore, one may expect an even higher conversion efficiency by using longer (picosecond or nanosecond) pulses and longer samples. For comparison, we also measured the SHG signal of the

LiNbO₃ waveguide without domain inversion (shown with red dots in Fig. 4). It can be seen that periodic poling leads to over 440 fold increase of the conversion efficiency. It is worth noting that the inscribed QPM domain patterns basically did not affect transmission characteristics of the waveguide. We have compared the output powers of the fundamental wave from the QPM and pure waveguides in an undepleted pump regime and found that an average propagation loss caused by the inscribed periodic domain patterns to be below 0.06 dB/cm, which is two orders of magnitude less than that measured in other femtosecond laser engineered QPM schemes [12, 13].

Fig. 4. The average power (a) and conversion efficiency (b) of second harmonic versus the average power of fundamental wave at the optimal quasi-phase matching temperature 62.5 °C. The black squares and red dots represent the results of quasi-phase matched and pure waveguides without poling, respectively. The inset depicts details of SHG in the latter case.

In conclusion, we have demonstrated all-optical fabrication of quasi-phase matched structures based on ferroelectric domain inversion in LiNbO₃ waveguides using femtosecond infrared pulses. The proposed scheme allows one to realize an efficient quasi-phase matching using the highest modulation depth of nonlinearity from $+\chi^{(2)}$ to $-\chi^{(2)}$, without introducing propagation loss for interacting waves. Conversion efficiency of 17.45 % is measured for the second harmonic generation in a 10 mm long domain inverted pattern. Our results indicate that the infrared laser poling constitutes a powerful method for fabricating periodic ferroelectric domains in an all-optical manner, thereby allowing for a wealth of new possibilities for precise and flexible domain engineering.

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