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Femtosecond laser writing of waveguides in periodically poled lithium niobate preserving the nonlinear coefficient

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Optical waveguides have been inscribed in periodically poled lithium niobate by femtosecond laser pulses with the multiscan technique. Second harmonic generation experiments from a fundamental wavelength of 1567 nm demonstrate that the nonlinear optical coefficient in the waveguides is preserved, yielding a conversion efficiency of 18% W^{-1}. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2748328]

Lithium niobate is one of the most important crystals for photonic applications. In fact, it has very high nonlinear and electro-optic coefficients that make it widely used for nonlinear devices and electro-optical modulators, both in academic research and in industrial products. In particular, periodically poled lithium niobate (PPLN) allows one to choose arbitrarily the phase matching wavelength and to exploit the highest nonlinear coefficient (d_{33}) for an efficient frequency conversion.[1,2] The fabrication of waveguides in lithium niobate is achieved mainly by two technologies: titanium indiffusion[1] and proton exchange.[2] Both techniques have provided excellent results in terms of waveguide quality and conversion efficiencies in nonlinear optical processes. They require, however, the use of photolithography to define channel waveguides and of a multistep process to either fabricate PPLN titanium-in-diffused waveguides or to anneal and bury the proton exchanged ones. Recently a great interest has arisen on the possibility to directly write optical waveguides in lithium niobate by means of femtosecond laser pulses.[3–9] This technology allows one to produce channel waveguides without photolithography and to directly bury them without subsequent fabrication steps. Therefore, femtosecond laser writing of waveguides could be a very flexible and low cost technology for the fabrication of integrated optical devices in lithium niobate with the added value of three-dimensional writing capability.

Since the first paper on this subject[3] several results have been achieved. Waveguiding at 1.55 μm (Refs. 4 and 5) and very low propagation[5,6] and insertion[6] losses have been demonstrated. Second harmonic generation (SHG) in femtosecond-laser-written waveguides has been performed exploiting both birefringence[7] and quasi-phase-matching,[8] and the electro-optic coefficient in the modified region has been measured.[9] However, several issues remain open. In particular, two key problems are degradation of the nonlinear coefficient in the irradiated region and waveguide stability both at room temperature and after annealing. In recent experiments on femtosecond-laser-written waveguides in PPLN,[8] the normalized SHG conversion efficiency was as low as 3×10^{-4}% W^{-1} cm^{-2}, clearly indicating that the nonlinear coefficient in the waveguide is almost completely canceled. On the other hand, experiments on SHG exploiting birefringence phase matching[7] show that the nonlinear coefficient in the femtosecond-laser-written waveguide can be preserved equal to that of the bulk, but the conversion efficiency was limited to 0.6% W^{-1} cm^{-2}; this is due to the low nonlinear coefficient (d_{15}) exploited in birefringence phase matching and to the high effective cross section (S_{eff}) obtained by overlapping a zero order mode at the fundamental and a first order mode at the second harmonic. The waveguide stability in time and temperature has been addressed in several papers. At room temperature, waveguide stability was observed to range from 1 month[4] to more than 2 months.[5] In a recent paper we demonstrated room temperature waveguide stability for more than 4 months.[6] On the other hand, heating the waveguides to more than 150 °C caused degradation in a few hours in all the experiments reported,[5,7,9] apart from some of the waveguides described in Ref. 7.

In this letter we report on frequency doubling of telecommunications radiation in PPLN waveguides fabricated by femtosecond laser pulses with the multiscan technique. The experimental results demonstrate a full preservation of the nonlinear coefficient and provide the highest normalized SHG conversion efficiency in femtosecond-laser-written waveguides.

Writing waveguides in lithium niobate with femtosecond laser pulses is much more critical than in glasses. In fact, modification of crystals requires a larger energy density. However, the high nonlinearity of lithium niobate (its third order susceptibility being almost ten times that of silica) and the strong nonlinear absorption associated with the extremely high peak intensity of such focused femtosecond pulses strongly affect the propagation of the pulse and the volume of the modified region. In particular, filamentation and multiple foci have been observed,[9] which prevent from controlling the position and the shape of the waveguide cross section. Lowering the pulse energy to avoid these effects provides faint waveguides that hardly show any guiding capability.[5,9] In order to deposit a sufficient energy for wave-

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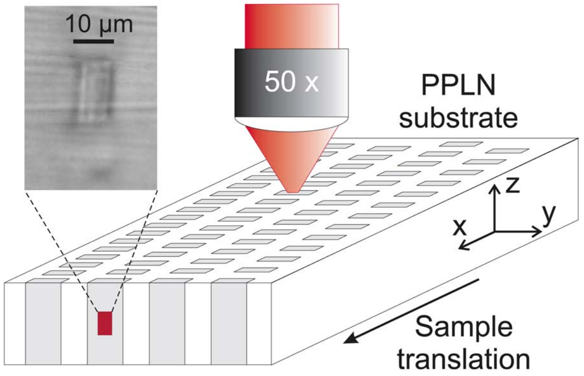

FIG. 1. (Color online) Schematic of the writing setup. Inset shows a microscope image of the waveguide end face.

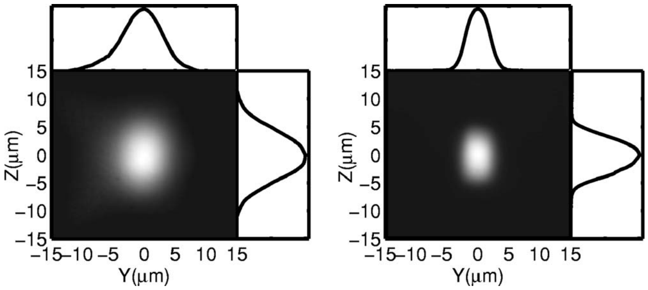

FIG. 2. Near field of the waveguide modes at (a) 1567 nm and (b) 783.5 nm. Lateral panels show the intensity profiles across the peak intensity.

guide fabrication, while keeping weak nonlinear interactions, different schemes have been proposed: (i) the pulse duration is increased up to several hundreds of femtoseconds,[4,5,9] (ii) a circular polarization is used,[5] and (iii) a multiscan approach is employed.[6,8] We believe that the multiscan approach is very favorable for writing waveguides in crystals since it allows one to create arbitrary cross-sectional shapes and sizes while keeping the single scan energy to a level that minimizes the nonlinear interactions. In addition, the creation of the guiding region right in the irradiated volume allows a superior control of the waveguide mode that could be mode matched to that of a fiber much more easily.

The waveguides were fabricated on a 20-mm-long *Z*-cut PPLN substrate (National Optics Institute, Quebec, Canada) that contained 12 different poling periods, from 18.35 to 20.9 μm. Figure 1 reports a schematic of the setup. The writing laser was a cavity-dumped Yb:KYW oscillator,[10] providing 350 fs pulses at 1030 nm with a repetition rate of 600 kHz. The laser linear polarization was set in the y direction. The femtosecond laser pulses were focused by a 50× objective [0.6 numerical aperture (NA)] at a depth of 250 μm from the surface. The explored pulse energies ranged from 170 to 470 nJ and processing speeds from 2 to 8 mm/s. Each waveguide was fabricated by 20 scans in the x direction, spaced by 0.4 μm in the y direction. The experimental setup for SHG experiments comprises a tunable laser (Agilent 8164B) with up to 7 mW output power in the range of 1440–1640 nm. The laser is coupled to a polarization maintaining fiber that is butt coupled to the PPLN waveguide. The input polarization is adjusted in order to excite TM modes which are the only ones supported by the femtosecond-laser-written waveguides. The output of the waveguide is collected by a 20× microscope objective (0.3 NA) and imaged onto a Vidicon and a charge coupled device camera for acquiring the intensity profiles and on Ge and Si photodetectors for fundamental and second harmonic power measurements.

Previous experiments on plain lithium niobate evidenced a fabrication parameter set (pulse energy of 370 nJ and writing speed of 4 mm/s), providing very good results in terms of waveguide insertion losses.[6] Such results were replicated on the PPLN sample. However, the SHG experiments evidenced a strong reduction of the nonlinear coefficient (1% of the original value). Further exploration of the parameter range evidenced a new window of parameters that yielded high quality waveguides with a much lower fluence. In particular, the best set was pulse energy of 270 nJ and writing speed of 6 mm/s. Two different mechanisms of waveguide formation are involved, corresponding to laser irradiation with high and low fluences. In the former case partial amorphization of the crystal occurs with stress-induced refractive index changes; however, strong lattice distortions spoil the nonlinear coefficient. In the latter case, the crystalline lattice is mainly preserved and the refractive index increase may be due to small lattice distortions and molar volume variations.[9] A microscope image of the end face of one of such waveguides is reported in the inset of Fig. 1. The sharp rectangular shape is due to the multiscan fabrication technique.

Propagation losses of 0.6 dB/cm and coupling losses of 1 dB/facet have been measured for the fundamental TM mode. The waveguide is single mode both at the fundamental and at the second harmonic as testified by the single peak observed in the second harmonic curve as a function of the pump wavelength. The near field profiles of the TM waveguide modes are reported in Fig. 2 both at 1567 nm (a) and at the generated second harmonic 783.5 nm (b). From the mode near fields, assuming a waveguide step-index profile with 8×16 μm^2 dimensions (see inset of Fig. 1), a refractive index change of $\Delta n \cong 3\times 10^{-3}$ is estimated. It is worth noting that the mode profiles show a limited ellipticity and are very symmetric in their shape as it can be observed in the lateral panels reporting the intensity profiles across the peak value. This result is a consequence of the fact that the waveguiding is achieved in the irradiated region which is very uniform and can be finely controlled in shape. The waveguide effective cross section (S_{eff}), calculated from the overlap of the two modes reported in Fig. 2, gives a value of 245 μm^2, which is very promising for an efficient frequency conversion. Further optimization, e.g., using a microscope objective with a slightly higher NA, could provide a fundamental waveguide mode which is matched to that of a telecommunications fiber, further reducing the insertion losses, and an even smaller waveguide effective cross section.

Figure 3 shows the room temperature second harmonic generation efficiency, normalized to the input pump power, as a function of the pump wavelength for a poling period $\Lambda = 19.2$ μm. A peak value of 18% W^{-1} is achieved for a fundamental wavelength of 1567 nm. To interpret the experimental results, the normalized SHG efficiency, $\bar{\eta} = P_{2\omega}/(P_{\omega})^2$, has been calculated by numerical integration of the coupled-mode equations:

$$\frac{dA_{\omega}}{dz} = -i\kappa d(z) A_{\omega}^{*} A_{2\omega} \exp(-i\Delta\beta z) - \alpha A_{\omega},$$

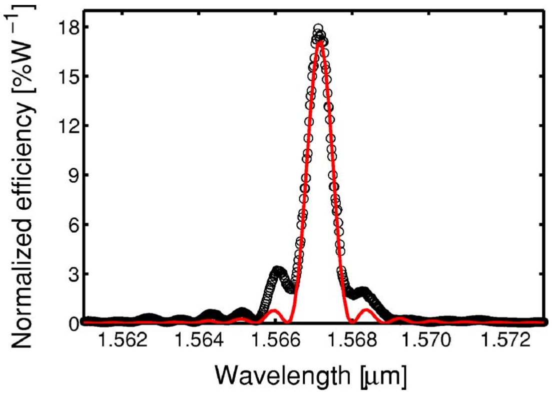

FIG. 3. (Color online) Normalized second harmonic generation efficiency as a function of pump wavelength: experimental data (circles) and theoretical fit (solid line).

$$\frac{dA_{2\omega}}{dz} = - i\kappa d(z)A_{\omega}^{2}\exp(i\Delta\beta z) - \alpha A_{2\omega},$$

$$\kappa^2 = \frac{2\omega^2}{\epsilon_0 c^3}\frac{1}{n_{\mathrm{eff},\omega}^{2} n_{\mathrm{eff},2\omega} S_{\mathrm{eff}}}, \tag{1}$$

where A_i is the field amplitude at the fundamental or second harmonic, normalized such as $|A_i|^2 = P_i$; $d(z) = d_{33}g(z)$ is the nonlinear coefficient in the waveguide region and $g(z)$ is a square function with period Λ; $n_{\mathrm{eff},i}$ is the effective index of the guided mode; S_{eff} is the effective waveguide cross section; $\Delta\beta = 4\pi(n_{\mathrm{eff},2\omega} - n_{\mathrm{eff},\omega})/\lambda$ is the propagation constant mismatch; and α is the propagation loss coefficient, which is assumed to be the same both at the fundamental and at the second harmonic. By integrating the above equations with L and d_{33} as free parameters, a very good fitting of the experimental results can be obtained (solid line in Fig. 3). From a physical point of view the two free parameters, L and d_{33}, are retrieved almost independently if the whole SHG efficiency curve is fitted.[11] In fact, L is determined by the width of the main peak and by the positions of the zeros of the curve, while d_{33} depends exclusively on the amplitude of the curve. The fitting value for the interaction length L is equal to 1.66 cm. This is quite close to the sample length of 2 cm but evidences that the waveguide uniformity can still be improved. Such slight disuniformity also explains why the two shoulders aside from the main peak are a little higher than the fit.[11] The nonlinear coefficient d_{33}, determined from the best fit, has a value of 21 pm/V. This value is exactly equal to that retrieved from a preliminary characterization of bulk SHG in the same sample, thus demonstrating that the femtosecond laser writing process with the above mentioned parameters is capable of preserving the material nonlinear coefficient and also the periodic poling. This allowed us to demonstrate a normalized SHG efficiency of $6.5\%\ \mathrm{W}^{-1}\,\mathrm{cm}^{-2}$, which is more than one order of magnitude higher than the best result published in the literature for femtosecond-laser-written waveguides.[7]

The waveguide stability in time and temperature has also been evaluated. At room temperature the stability is very good, showing constant performances after 2 month testing. On the other hand, a thermal treatment at 150 °C for 4 h caused a consistent lowering of the confining capabilities of the waveguide with a consequent increase of the effective waveguide cross section S_{eff} and a decrease of the SHG efficiency by one order of magnitude. This problem is common to the previous literature[4,5,9] apart from waveguides presented in Ref. 7, where, however, the waveguiding region was not directly affected by the femtosecond laser. In all other cases, there seems to be a trade-off between stability at high temperature and preservation of the nonlinear coefficient. In fact, waveguides written with higher pulse energy remained unchanged after the thermal treatment but provided a nonlinear coefficient decreased by a factor of 100.

In conclusion, we have demonstrated that femtosecond laser writing of waveguides in PPLN can preserve the nonlinear coefficient and the periodic poling, providing a SHG efficiency of $18\%\ \mathrm{W}^{-1}$. This result paves the way to femtosecond laser writing of waveguides in nonlinear crystals for efficient frequency conversion by exploiting the quasi-phase-matching technique. In particular, crystals as periodically poled potassium titanyl phosphate or lithium tantalate can be used for high power nonlinear frequency conversion at room temperature, due to a higher optical damage threshold; this could overcome the limitation of high temperature stability of such waveguides. Experiments on femtosecond laser writing of waveguides in these materials are currently in progress.

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