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Research Article

Formation of $MgO:LiNbO_3$ Domain-Inverted Gratings by Voltage Application under UV Light Irradiation at Room Temperature

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$MgO:LiNbO_3$ is an attractive nonlinear-optic crystal for quasiphasematched (QPM) nonlinear-optic devices. This paper reports a new formation method of domain-inverted gratings for QPM in $MgO:LiNbO_3$. Domain inversion of $MgO:LiNbO_3$ by voltage application under UV light was characterized, and reduction of the voltage required for inversion was demonstrated. Results of voltage application under periodic UV light suggested that suppression of excess lateral expansion of the domain inverted regions on $-Z$ surface was crucial for domain-inverted grating formation. Voltage application to a crystal with a photoconductive cladding layer under periodic UV light was proposed. The cladding layer suppressed the expansion, and the domain-inverted gratings with period of 18 μm and area of 25×5 mm^2 were obtained. The formation method does not require the photolithography process and allows the formation by voltage application at room temperature, and therefore, is quite simple and productive.

1. Introduction

There has been a great interest in quasiphasematched (QPM) nonlinear-optic devices for applications in variety of fields including optical memory, optical communication, display, and quantum optics, because they can provide efficient wavelength conversion through the largest nonlinear-optic tensor element at any wavelength in the transparency range [1]. Ferroelectric-domain-inverted grating in nonlinear optic crystals is the mostly used structure to accomplish QPM. Formation technique of the grating by voltage application through periodic electrodes has been established for $LiNbO_3$ crystal [2–4], and a lot of QPM devices have been demonstrated in $LiNbO_3$ [1]. However, applications of the devices may be limited due to the photorefractive damage caused by intense short wavelength optical waves. The devices are often kept at high temperature during the experiments to avoid the damage.

MgO-doped $LiNbO_3$ has large nonlinear-optic coefficients comparable to those of the nondoped $LiNbO_3$ and higher resistance to the photorefractive damage [5], and therefore, it is an attractive nonlinear-optic crystal for QPM devices. Domain-inversion characteristics of $MgO:LiNbO_3$ are quite different from those of the nondoped $LiNbO_3$ [6–8]. Formation of domain-inverted gratings in $MgO:LiNbO_3$ has been studied extensively, and several methods have been reported [9–13]. These methods require photolithography process for fabrication of periodic electrodes and also special treatments for voltage application, such as corona discharge, crystal heating, insulation layer cladding, and/or vacuum environment. Simpler and more productive methods are preferable for practical application.

We recently found reduction of voltage required for $MgO:LiNbO_3$ domain inversion by ultraviolet (UV) light irradiation and showed possibility of grating formation at room temperature by a simple method without photolithography process [14]. The reduction was confirmed also by another research group [15].

In this paper, we report the characteristics of the domain inversion under UV in detail, and demonstrate formation of domain-inverted gratings in $MgO:LiNbO_3$.

2. Reduction of Inversion Voltage by UV Irradiation

We characterized domain inversion of Z-cut MgO(5mol%):$LiNbO_3$ by voltage application under UV light irradiation

using a setup shown in Figure 1. On the $+Z$ face of a $MgO:LiNbO_3$ crystal of 0.5 mm thickness a NESA glass plate was settled, and LiCl aqueous solution filled the gap between the plate and the crystal. The solution served as a liquid electrode. The $-Z$ face was grounded through a liquid electrode and a series capacitor for monitoring the transferred charge. The area of the electrodes was about 4 mm^2. An ultra-high-pressure mercury lamp was used to irradiate the crystal through the NESA. The intensity of the light was ~0.1 W/cm^2 on the NESA. Voltage of a single cycle symmetric triangular waveform was applied to the crystal at room temperature. The amplitude was ~3 kV and the period was 10 seconds. Recording the transferred charge during the voltage application, we obtained a hysteresis loop as shown in Figure 2(a), which implies domain inversion. Voltage application was stopped at point A in Figure 2, and the crystal was etched in $HF:HNO_3$ mixture to visualize the domain structure. Inversion of domain was confirmed. For application of the identical voltage without the UV irradiation, the charge transfer was much smaller, as shown in Figure 2(b), and no domain inversion took place. The results indicated that UV light irradiation reduces the voltage required for domain inversion.

By application of a voltage of −2 kV for 4 seconds to the $-Z$ surface with irradiating the UV light of ~0.1 W/cm^2 intensity from $-Z$ side, domain-inverted region was obtained only on $-Z$ surface. By application of a voltage of +2 kV for 4 seconds to $+Z$ surface with irradiating from $+Z$ side, inverted region was not obtained on $+Z$ surface but on $-Z$ surface. This result suggests that domain inversion nucleates only on $-Z$ face under the UV irradiation. It is quite different from the other methods, where inversion nucleates on $+Z$ face.

The light from the high-pressure mercury lamp consists of many emission lines, and they include the emissions at 289, 313, 334, and 365 nm wavelengths. Each wavelength component was extracted by a wavelength filter, and used for the domain inversion experiments to clarify the wavelength components effective to the reduction of the inversion voltage. The inversion took place for UV irradiation at 289 or 313 nm wavelength, which is in the absorption wavelength range of $MgO:LiNbO_3$ crystal. The mechanism of the UV-assisted domain inversion is not clear yet. It may be related to local field due to migration of free electrons caused by UV light absorption.

3. Domain Inversion by Voltage Application under Periodic UV Light

We considered that voltage application under UV light with spatially periodic intensity modulation would result in formation of $MgO:LiNbO_3$ domain-inverted grating. We made an apparatus for the voltage application at room temperature under periodic UV light. On the $-Z$ surface of a $MgO:LiNbO_3$, a Cr photomask with grating pattern is settled. A UV light from an ultra-high-pressure mercury lamp goes through the photomask, and periodic intensity distribution of UV light is formed in the crystal. LiCl aqueous solution fills the gaps on both surfaces of the crystal. A negative high voltage is applied to the $-Z$ surface at room temperature through the uniform liquid electrode. Domain

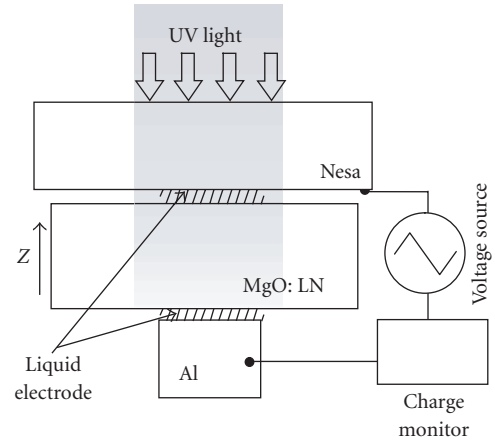

Figure 1: Setup for characterization of domain inversion under UV irradiation.

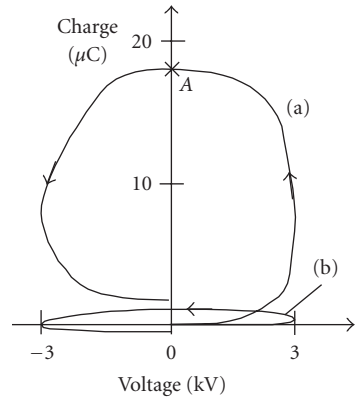

Figure 2: Hysteresis loop of $MgO:LiNbO_3$ crystal at room temperature (a) under UV irradiation and (b) without UV irradiation.

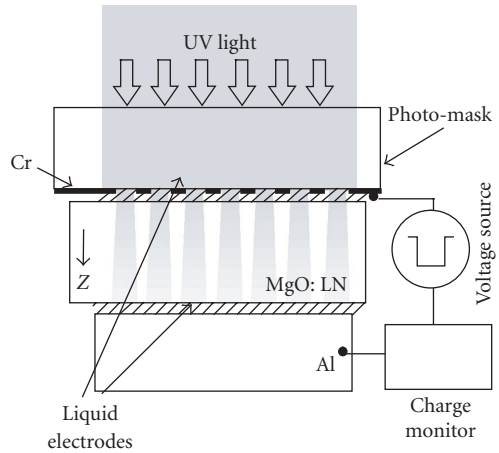

Figure 3: Setup for voltage application under spatially periodic UV light at room temperature.

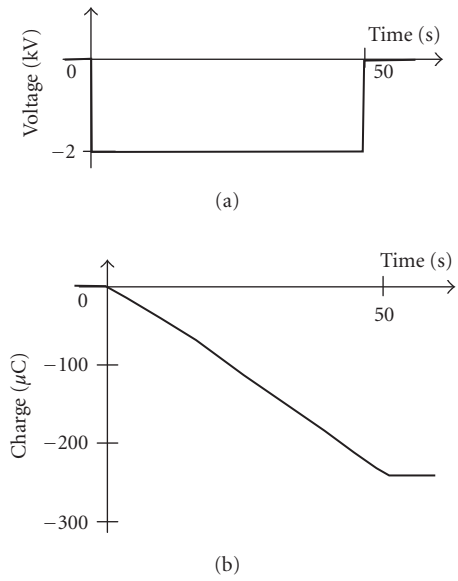

Figure 4: Typical waveforms of the applied voltage and the accumulated inversion charge.

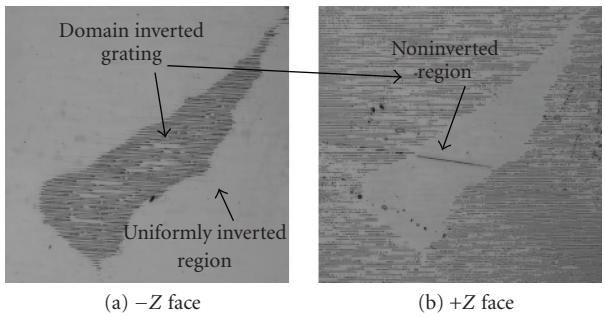

Figure 5: Typical domain structure formed by voltage application under periodic UV irradiation.

inversion takes place in the UV irradiated region, while no inversion takes place in the nonirradiated region. Therefore domain-inverted gratings are formed.

Voltages were applied under UV light of 0.1 W/cm^2 from $-Z$ side, and the resultant domain structures were examined after etching. The crystal thickness was 0.5 mm, and the period of the Cr pattern was 15.4 μm. The applied voltages ranged from -0.2 to -4 kV, and the durations ranged from 10 to 90 seconds. Typical waveforms of the applied voltage and the accumulated inversion charge are shown in Figure 4 for electrode area of 50 mm^2. The charge increased almost proportionally. Typical result is shown in Figure 5. In a part (central part of Figures 5(a) and 5(b)), grating was obtained on $-Z$ face, but inverted region was not obtained on $+Z$ face yet. In the surrounding part, inverted regions had expanded laterally and merged one another on $-Z$ face. Only in such part, inverted regions were obtained on +Z. Nucleation and evolution of domain-inverted regions on the $-Z$ surfaces and on the cross sections in the vicinity of the $-Z$ surfaces were examined and are shown in Figure 6. At first domain-inverted regions nucleated at the $-Z$ surface (Figures 6(a) and 6(b)). The initially-domain-inverted regions expanded for higher and longer voltage application, and a domain-inverted grating was formed on the $-Z$ surface (Figure 6(c)). The inverted region was limited in the vicinity of $-Z$ surface. The depth of the inverted region was a few microns (Figure 6(d)). The regions laterally expanded at the $-Z$ surface and formed a very thin domain-inverted layer (Figures 6(e) and 6(f)). The thickness of the layer was $\sim$0.5 μm. Besides, next the initially-inverted regions started expansion toward $+Z$ surface to form immediately a domain-inverted grating with inverted regions continuing in the whole crystal thickness (Figures 6(g) and 6(h)). Further voltage application resulted in a uniformly inverted structure without grating. The domain inversion took place in the same way under UV irradiation of higher optical intensity up to 2 W/cm^2. Suppression of excess lateral expansion of domain-inverted region on $-Z$ surface is crucial for forming domain-inverted gratings applicable to QPM devices.

4. Formation of Domain-Inverted Gratings

We modified the method to suppress the excess lateral expansion. Voltage pulse was applied to a crystal with photoconductive cladding layer on $-Z$ surface under UV. The application setup was similar to Figure 3 except that a photoconductive layer was deposited on the $-Z$ surface. In the UV-irradiated region, the photoconductive layer serves as a (semi-)conductor, allows charge transfer associated with domain inversion, and therefore, does not prevent domain inversion. On the other hand, in the nonirradiated region, it serves as an insulator and prevents inversion. Therefore, excess lateral expansion of the inverted region can be suppressed, and the grating structure can be maintained on $-Z$ surface until the inverted regions reach $+Z$ surface.

We noticed that a commercially available photoresist (Clariant, AZ-P4620) can be used as a photoconductive material for this purpose. The resistivity of the film was measured. It was initially $\sim 6 \times 10^{15}$ (Ωcm), and dropped down to $\sim 1 \times 10^{12}$ (Ωcm) by irradiation of UV light with intensity of 1 W/cm^2.

The photoresist was spin-coated on $-Z$ surface of MgO:LiNbO$_3$ crystal of 0.5 mm thickness, and voltage pulses were applied under various conditions. The thickness of the photoconductive layer, intensity of the UV light, duration of the voltage application, and the voltage were chosen within ranges of 7–10 μm, 0.05–0.5 W/cm^2, 50–1000 seconds, and 3.3–3.9 kV, respectively. The resultant domain structure was examined after etching. When the thickness was too thin and/or the intensity was too high, suppression of the lateral expansion was not sufficient. When the thickness was too thick and/or the intensity was too low, the width of the inverted region was too narrow. When the duration was too short and/or the voltage was too low, the inverted region did not reach to $+Z$ face. When the duration was too long and/or the voltage was too high, damages took place in the crystal. A domain-inverted grating fabricated for the photoconductive layer thickness of 10 μm, the UV intensity of 0.07 W/cm^2, the duration of 900 seconds, and the voltage of 3.5 kV, are

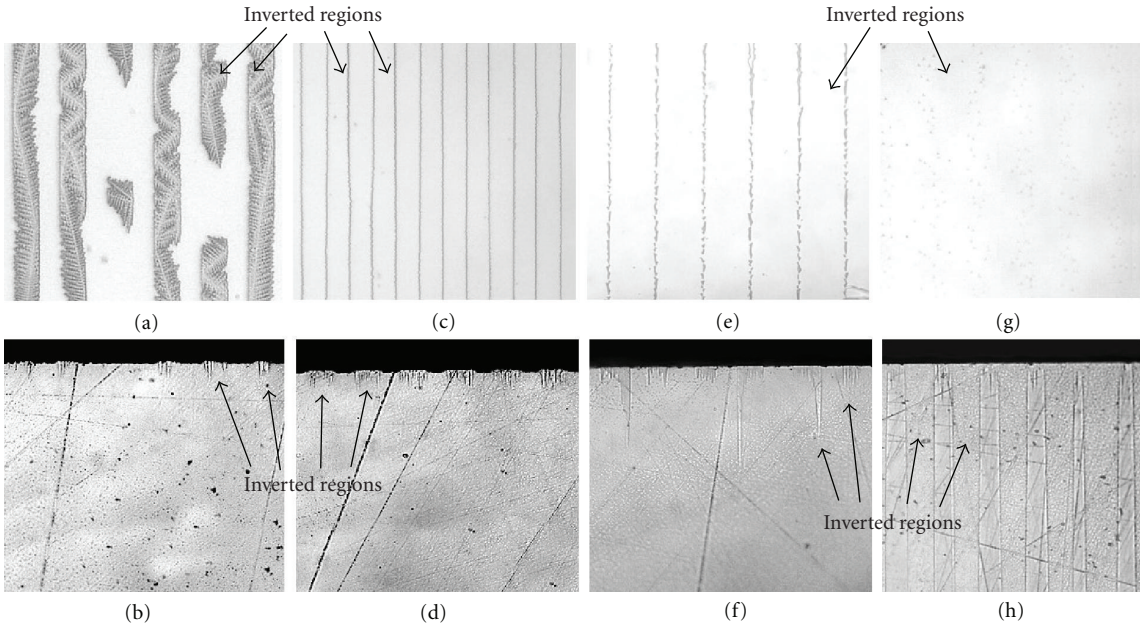

Figure 6: Nucleation and evolution of domain-inverted regions (a), (c), (e), (g) on $-Z$ surfaces and (b), (d), (f), (h) in the cross sections. Inverted regions evolved as ((a), (b))→((c), (d)) →((e), (f)) →((g), (h)). The period of the grating pattern was 15.4 μm.

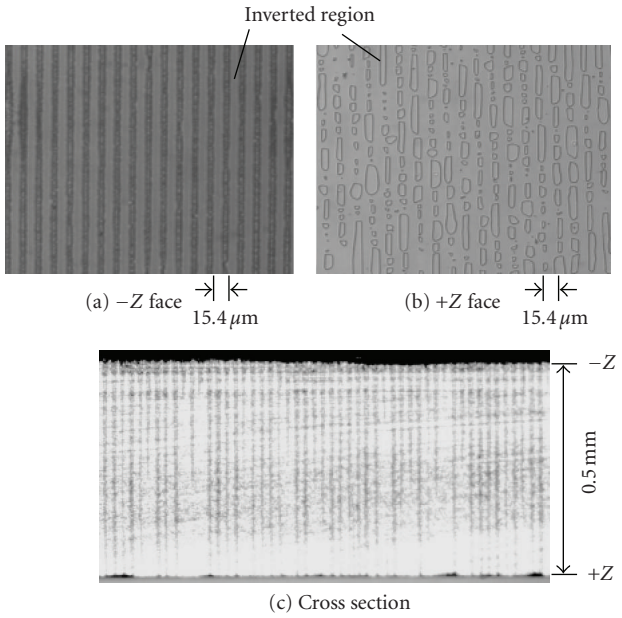

Figure 7: Domain-inverted grating formed by voltage application to crystals with photoconductive cladding under UV irradiation.

shown in Figure 7. The grating period was 15.4 μm and the area was $10 \times 5\,\text{mm}^2$. Grating structure was obtained on both $-Z$ face (Figure 7(a)) and $+Z$ face (Figure 7(b)) in the same part. The crystal was cut by a dicing saw and etched again to examine the domain structure on the cross section. The inverted regions continued through the whole crystal thickness (Figure 7(c)). However, there are some points to be improved further. They include that the width of the grating line on $-Z$ was somewhat wide, and that the grating lines on $+Z$ were not continuous lines but segmented lines.

Fabrication of domain-inverted gratings in larger area was carried out. A grating with period of 18 μm and area of $25 \times 5\,\text{mm}^2$ was obtained. In a simple bulk SHG experiment using the crystal, a QPM SH wave was obtained for a pump wavelength at 1549.5 nm. It agrees well with theoretical prediction based on a reported Sellmeier equation [1]. The SHG performance will be characterized in detail and reported elsewhere.

5. Conclusion

Domain inversion of MgO:$LiNbO_3$ by voltage application under UV light was characterized, and reduction of the inversion voltage was shown. Voltage application under UV light with spatial periodic intensity modulation suggested that it was crucial to suppress excess lateral expansion on $-Z$ surface for domain-inverted grating formation. Application of photoconductive cladding layer on the crystal was proposed and successfully suppressed the expansion. Domain-inverted gratings with period of 18 μm and area of $25 \times 5\,\text{mm}^2$ were obtained. The formation method does not require the photo-lithography process and allows voltage application at room temperature; and therefore it is quite simple and productive. Experimental work is being continued for higher-quality grating formation and for application of the gratings to QPM nonlinear-optic devices.

Acknowledgment

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