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Domain patterning in lithium niobate using spontaneous backswitching

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ABSTRACT

In nonlinear optics applications employing quasi-phasematching (QPM), short pitch domain gratings are generally required for the generation of visible and ultraviolet light. The conventional electric-field poling method enables the fabrication of periodically-poled lithium niobate (PPLN) down to generally 6 micron-pitch domains in 0.5-mm-thick substrates. While such PPLN is useful for first-order second harmonic generation (SHG) of green wavelengths, shorter periods for blue and UV SHG have been difficult to obtain in 0.5-mm-thick substrates. Here we describe an enhanced electric-field poling technique for ferroelectric materials which utilizes spontaneous flip-back towards high-resolution and high-yield domain patterning.

Keywords: Periodically-poled, lithium niobate, second harmonic generation, blue light, backswitching

1. INTRODUCTION

Quasi-phasematching (QPM) nonlinear optical processes entailing the conversion of visible and ultraviolet light typically require short-period domain gratings. In lithium niobate, domain periods of roughly 6 μm and shorter are generally useful for first-order QPM second harmonic generation (SHG) of green, blue and ultraviolet wavelengths. A conventional electric field poling technique is well established for the fabrication of periodically poled lithium niobate (PPLN) with periods down to 6.5-μm[1], and has been applied to other ferroelectric materials including $LiTaO_3$[2] and $KTiOPO_4$ (KTP)[3]. Because of the large effective nonlinear optical coefficient of PPLN, $d_{eff} \approx 21$ pm/V, it continues to attract interest as a QPM material for the generation of blue and ultraviolet wavelengths[4]. Recent demonstrations of blue light SHG in 4.5-μm-period PPLN include 5 mW from a 102-%/W-efficient waveguide pumped at 967 nm by a tunable diode laser[5], and 450 mW at 40 % efficiency pumped by a pulsed 946 nm Nd:YAG laser[6]. Nonetheless, utilization of the conventional poling technique at this and shorter QPM periods has been limited by a reduced d_{eff} due to uncontrolled spreading and merging of domains in the bulk material.

2. BULK DOMAIN PATTERNING

Lithium niobate, $LiNbO_3$, is an attractive material for quasi-phasematching nonlinear optics applications. The use of an applied electric field at room temperature towards the control of domain inversion in the bulk, by Yamada et. al.[7], initiated significant research toward obtaining periodically-poled $LiNbO_3$ (PPLN). The result of this work has been the standardization of the 'conventional poling method'[8].

2.1. The poling set-up

The conventional poling method generally begins with $LiNbO_3$ wafer substrates of congruent composition. The single domain 0.2-0.5 mm thick samples are photolithographically patterned with a periodic electrode structure deposited on z+ surface[7,8] The electrodes may consist of either patterned metal lines or trenches in photoresist. For metal electrodes, a thin insulating overcoat layer is applied in order to inhibit growth of the domains between the electrodes (Fig. 1).

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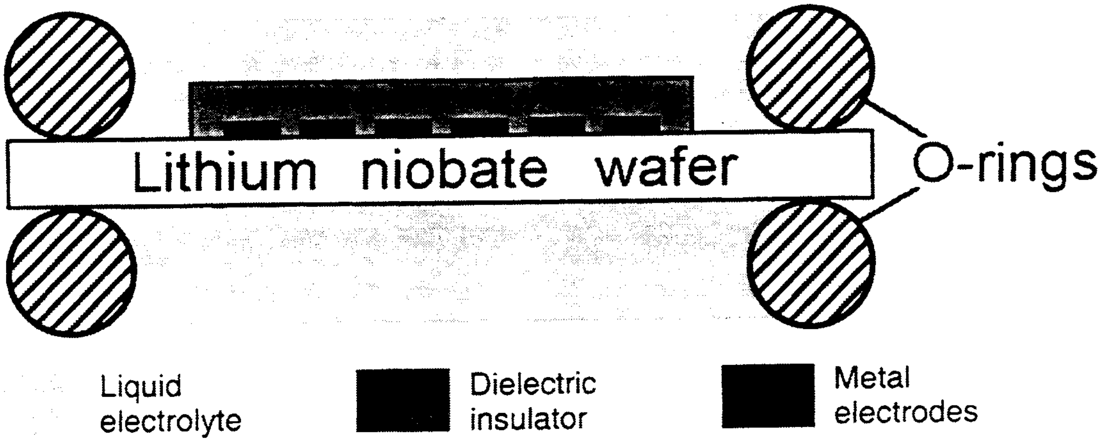

Figure 1. Schematic diagram of the conventional electric field poling fixture.

An external electric field is applied to the sample by electrical contact with the patterned electrodes and unpatterned wafer surface via the liquid electrolyte contained in the poling fixture. A waveform generator and high voltage amplifier are used to provide the poling pulse; both the voltage and current are monitored during the poling. The parameters of the domain structure are controlled by the pulse shape and current value. After complete or partial poling the polar surfaces and cross-sections are etched for 5-10 minutes in hydrofluoric acid at room temperature. The surface relief associated with domain patterns is visualized by both optical and scanning electron microscopes. Visualization of the domain structure, along the crystal x-direction, can be enhanced by choosing appropriate tilted cross-sections. The comparison of domain patterns obtained for different durations of poling pulses yields information about domain evolution.

2.2. Stages of the domain evolution

Analysis of the domain patterns after partial poling reveals several stages of domain evolution[8,9,10]. The poling process starts with "nucleation" (arising of new domains) at z+ polar surface along the electrode edges when the polar component of the local field E_z exceeds the threshold value[8,9,11] (Fig. 2a). The second stage represents forward and sideways growth and merging of the domains under the electrodes[9] (Fig. 2b). At the third stage, planar walls of formed laminar domains move out, away from the electrodes[9] (Fig. 2c). After terminating the switching process by rapidly lowering the poling field, two possibilities exist: either the stabilization of the generated domain structure, or the partial backswitching ("flip-back") of domains to the initial state. The stabilization of the domain structures in the conventional poling method requires a separate stage in the poling voltage waveform specifically for the purpose of suppressing the backswitching process after removing the poling field (Fig. 3a)[8,9,12,13].

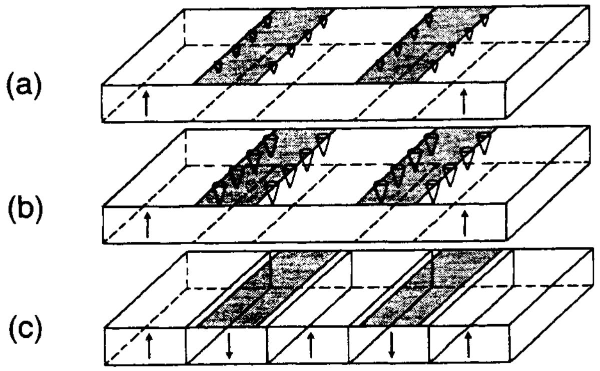

Figure 2. The main stages of domain evolution during switching in single domain plate with stripe electrodes: a) nucleation at electrode edges; b) growth and merging of domains beneath the electrodes; and c) laminar domains move out past electrode edges.

2.3. Backswitching

Spontaneous flip-back, or backswitching, of inverted domains is well known in the field of ferroelectric materials engineering[14]. For many ferroelectric materials, if the external electric field is suddenly removed during or immediately after poling, the poled domains tend to backswitch. As shown in Figure 3, this behavior is also demonstrated in the fabrication of photolithographically patterned ferroelectrics such as PPLN and periodically poled $LiTaO_3$ (PPLT). In these cases where the external electric field was suddenly removed once the domain duty cycle had reached approximately 50%, backswitched domains nucleated along the electrode edges. The high density and submicron spacing of these backswitched nuclei led us to investigate the utilization of controlled backswitching as a means for achieving high fidelity ferroelectric domain patterning[15].

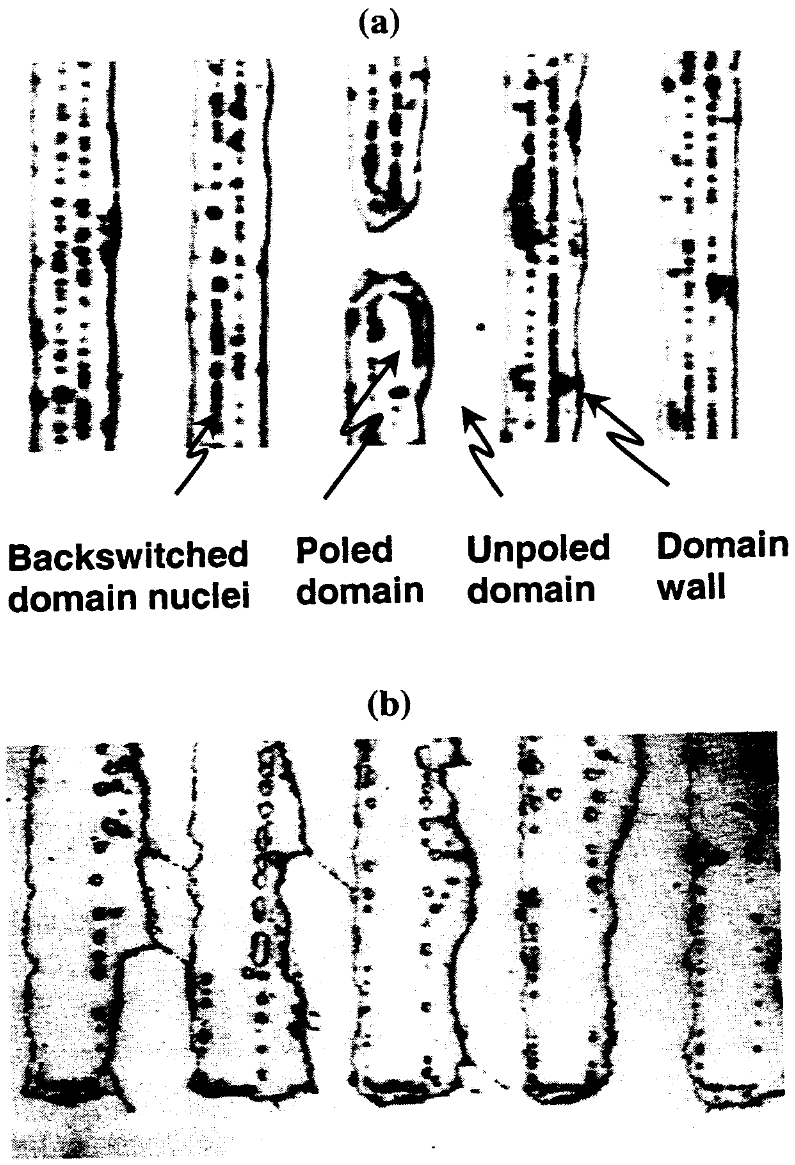

Figure 1. Top (+z) surface images of backswitched domain nucleation along electrode edges in (a) 10-μm-period PPLN, and (b) 26-μm-period PPLT.

3. POLING WAVEFORMS

Here we describe typical voltage waveforms for both the conventional and backswitch electric field poling methods.

3.1. The conventional poling waveform

A typical conventional poling field waveform, implemented for a photolithographically patterned 76.2-mm-diameter $LiNbO_3$ wafer, is shown in Figure 3a. The conventional poling pulse begins with a ramp up to an approximately 0.4-ms-long spike at 24 kV/mm during which domains are nucleated along the edges of the electrodes. The nucleated domains are then grown out at the coercive field $E_c \approx 21.75$ kV/mm. During this "forward growth" stage, the domains are grown out past the electrodes, and under the insulator. After forward growth duration of approximately 73 ms, the domain spatial duty cycle will approach 50%. The external field is then lowered to a stabilization field[8,9] of approximately 19.5 kV/mm for a duration of 10 ms in order to allow screening of the depolarization field by free charges. Ramping the external field to 0 kV/mm over a duration of 60 ms then terminates conventional poling.

3.2. The backswitch poling waveform

A backswitch poling waveform, also implemented for a photolithographically patterned 76.2-mm-diameter $LiNbO_3$ wafer, is illustrated in Figure 3b. The backswitch poling waveform is identical to the conventional waveform from the start through to the forward growth stage. Unlike conventional poling, however, the E_c stage is sustained until forward growth is finished and the domains have fully spread beneath the insulator and merged in the bulk, reaching a duty cycle of approximately 100%. Upon completion of forward growth, the external field is lowered. During this "backswitching stage", the external field is decreased before the depolarization field has been screened by free charges in both the bulk material and external circuit. Therefore, the depolarization field, having switched sign due to the process of forward domain inversion, is momentarily large enough to cause the erasure, or backswitching, of the forward-switched domains. Depending on the speed of the screening processes and amplitude of the backswitching field, flip-back can continue until the domains have substantially returned to their original state. However, raising the external field back up to approximately 3.75 kV/mm can terminate backswitching. The ability to selectively initiate, maintain and terminate backswitching allows for the formation of domain patterns with small feature sizes and high uniformity through large volumes of material.

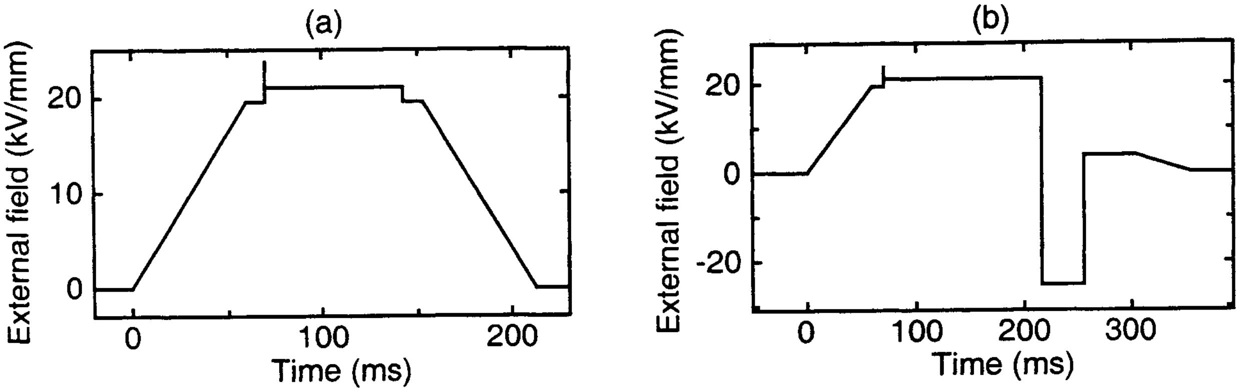

Figure 3. (a) Conventional poling electric field waveform; and (b) backswitch poling waveform .

4. BACKSWITCH POLING RESULTS

To demonstrate backswitch poling, we patterned 3-inch-diameter, 0.5-mm-thick lithium niobate wafers with 4–μm-period, 0.75–μm-wide, 750-Å-thick NiCr electrodes[1] on the +z face. In order to electrically insulate the NiCr gratings during poling, the patterned surface was overcoated with a 0.5–μm-thick patterned layer of photoresist and baked for one hour on a hotplate at 150 °C. To provide electrical contact to the NiCr gratings, the photoresist was patterned with an array of 100–μm-wide stripe-shaped openings. A photoresist stripe opening ran along the middle of each grating section.

4.1. Four-μm-period domain structures

The 4-μm-period patterned wafers were forward poled for 145 ms and then backswitched for 39.2 ms. Devices up to 50 mm in length were taken from the backswitch poled wafers. Figure 4 shows -z and y face views of the domain structures.

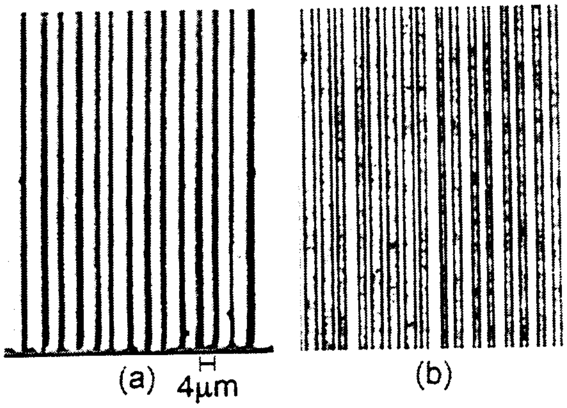

Figure 4. Images of backswitch-poled 4-mm-period domains in 0.5-mm-thick PPLN; (a) y face, and (b) –z face.

4.2. Spatial frequency multiplication

Backswitch poling also enables the formation of domain periods at multiples of the spatial frequency of the electrode structures. This can be accomplished by means of variations in both the electrode duty cycle and poling voltage waveform. Figure 5a is a $\mathbf{z}^+$ surface image, which demonstrates the possibility of spatial frequency doubling of 10–μm-period domains. In this case, the domain depth is typically 50-100 μm for 1 μm domain widths (Fig. 5b). Further, higher frequency multiplication of the domain pattern can be achieved in this fashion. Figure 5c shows the potential for spatial frequency tripling of 10-μm-period domains on the $\mathbf{z}^+$ surface. In this case, the additional sub-μm-wide domains arise just under the electrode edges and penetrate 20-50 μm deep (Fig. 5d).

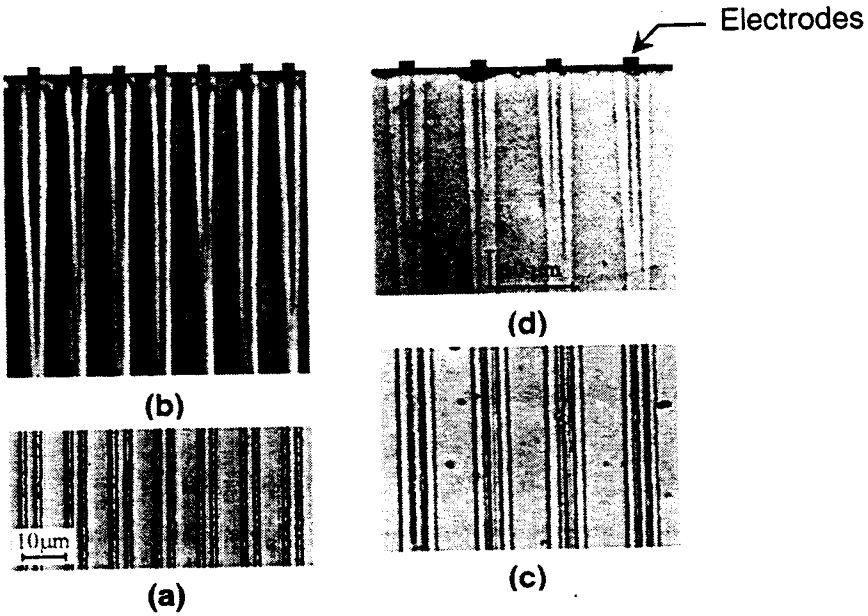

Figure 5. Spatial frequency multiplication of domain structures by backswitch poling in a sample having a 10 μm electrode period. Spatial frequency doubling: (a) z^+ surface; and (b) y surface view. Spatial frequency tripling: (c) z^+ surface; and (d) y surface view. In (d), the higher magnification in the x direction was obtained by using a tilted cross-section.

4.3. Erasing and splitting

Detailed analysis of the individual domain behavior during backswitching reveals two distinct variants of their evolution: erasing and splitting. The erasing process leads to formation of the backswitched domains in earlier switched area without any variation of the external shape of switched domain (Fig. 6a). During splitting the growing backswitched domain cuts the initial switched domain conserving its volume (Fig. 6b).

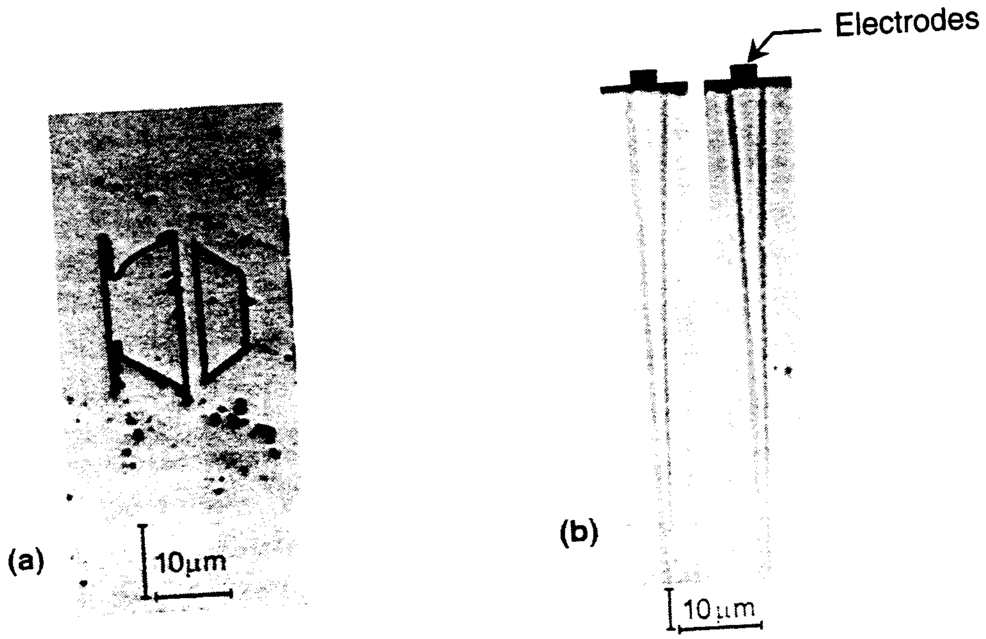

Figure 6. Domain changing by backswitching: (a) erasing (z+ view) and (b) splitting (y surface).

OPTICAL RESULTS

Uncoated 50-mm-length samples of the 4-μm-period backswitched PPLN samples were characterized by cw single-pass SHG of blue light using a Ti:S pump laser. The Ti:S laser was tuned to approximately 930 nm and focused to a 44 μm waist radius in the center of the samples. The crystals were heated to a corresponding phasematching temperature of approximately 280 °C. This phasematching temperature was chosen to avoid photorefractive effects and to reduce optical absorption in the sample due to the presence of blue light. Figure 7 shows plots of internal SHG power as a function of internal fundamental power. With 1 W of fundamental power, 60 mW of blue (465 nm) SHG was produced, giving a quadratic slope conversion efficiency of 6.1 %/W. The effective nonlinear coefficient $d_{eff} \approx 9$ pm/V (roughly half of the ideal value), for the 50-mm-length samples, was limited by a combination of variation in positions of the domain walls and thermal loading due to optical absorption. Figure 8 shows an SHG temperature tuning curve for a 50-mm-length backswitched PPLN sample using the Ti:S pump tuned to 920 nm with loose (100 μm waist radius) focusing.

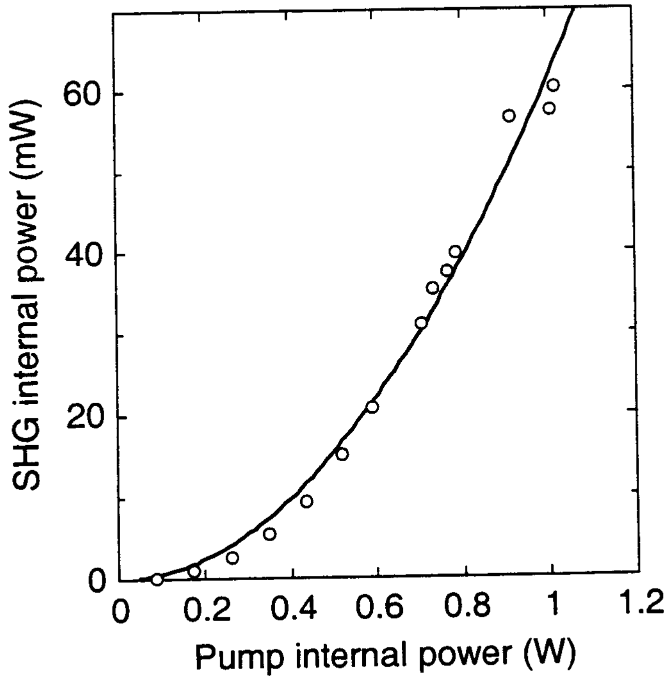

Figure 7. Plot of 465 nm internal cw single-pass SHG power vs. 930 nm Ti:S internal fundamental power

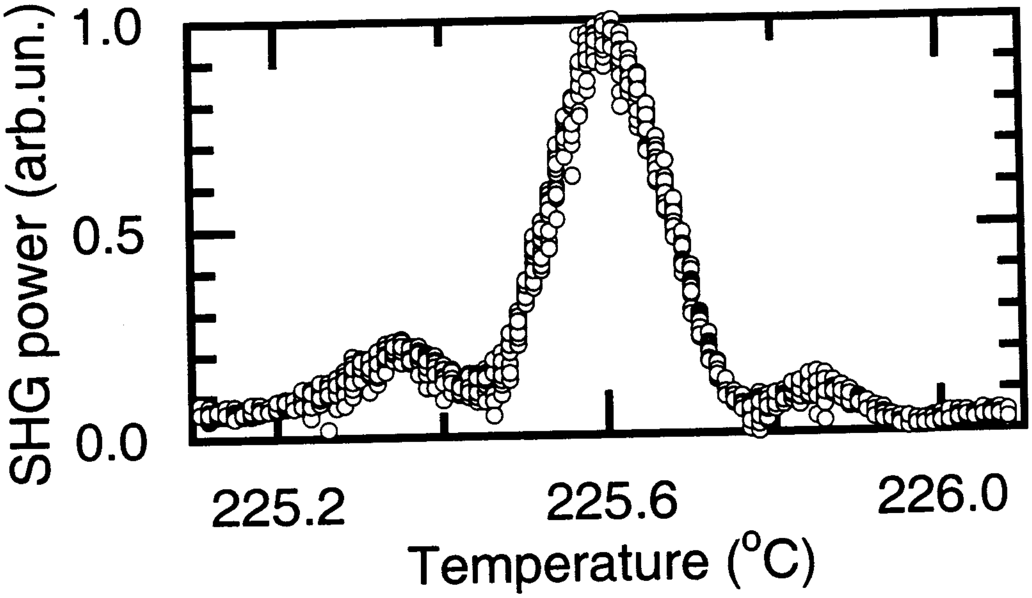

Figure 8. A temperature tuning curve for a 4–μm-period, 50-mm-length, 0.5-mm-thick backswitched PPLN sample (920 nm Ti:S pump).

In summary, backswitch poling has been investigated as an effective technique for achieving short-period wafer-scale patterning of 0.5-mm-thick $LiNbO_3$. Using 50-mm-length 4-µm-period samples, single-pass cw SHG at 465 nm produced 61 mW at 6.1-%/W-efficiency by a Ti:S laser source. Future work will include improving backswitch poling to obtain near-ideal effective nonlinearity over large sample areas, and applying the technique to other ferroelectric materials with lower optical absorption such as $LiTaO_3$.

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