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Domain switching characteristics and fabrication of periodically poled potassium niobate for second-harmonic generation

Joong Hyun Kim and Choon Sup Yoon^{a)}

Department of Physics, KAIST, Daeduck Science Town, Taejeon 305-701, Korea

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We report on the 180° domain switching characteristics and fabrication of periodically poled KNbO₃ for second-harmonic generation. Using a modified triangular pulse, a periodically poled KNbO₃ of very high quality was fabricated with dimensions of 3×4×0.925 (*a*×*b*×*c*) mm³ and a period of 32.5 μm to utilize the *d*₃₁ component of the second-order nonlinear optical coefficient tensor. An efficient second-harmonic generation of a mode-locked Nd:YAG laser, operated at 1064 nm with a pulse width of 35 ps, was achieved with a maximum conversion efficiency of 34.8%. The effective nonlinear coefficient, measured in the limit of undepleted pump approximation, was 5.2 pm/V, which is 68% of the theoretical value. © 2002 American Institute of Physics.
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Periodically domain inverted ferroelectric nonlinear optical crystals are ideal for efficient frequency conversion utilizing quasi-phase-matching (QPM).^{1,2} Lithium niobate,³ lithium tantalate,⁴ potassium titanyl phosphate^{5,6} (KTP), and its isomorph rubidium titanyl arsenate⁷ (RTA) are among the materials investigated the most. Each of these has its own merits and drawbacks. Although periodically poled lithium niobate (PPLN) and lithium tantalate have large effective nonlinear optical coefficients, 17 and 16.6 pm/V, respectively, they must be used at elevated temperatures because of a photorefractive effect at room temperature. Magnesium doping in lithium niobate can suppress the photorefractive effect, but the LiNbO₃: Mg requires an advanced poling technique. KTP and RTA can be operated at room temperature and can be poled as thick as 3 mm. However, since KTP has high electrical conductivity, poling should be done at low temperature⁵ or with K⁺ ions exchanged by Rb⁺ ions⁶ to reduce the conductivity. The conductivity of RTA is smaller than that of KTP by five orders of magnitude,⁸ but it has ferroelastic domains at room temperature. Consequently, RTA has to be made into a single domain before poling.

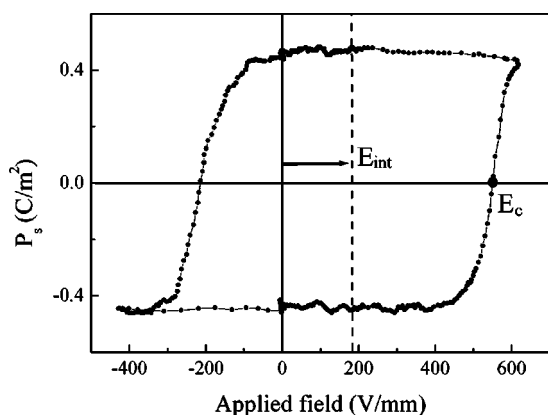
Periodically poled potassium niobate, KNbO₃ (PPKN) is a good candidate for nonlinear frequency conversion using QPM, due to its large nonlinear optical coefficient (*d*₃₃ = 20.6 pm/V), wide transparency range (0.4~5 μm) and freedom from photorefractive effects. In addition, a relatively low coercive field of KNbO₃ (~550 V/mm) makes it easy to fabricate PPKN. The fabrication of PPKN and second-harmonic generation (SHG) was first reported by Meyn *et al.*⁹ However, fundamental parameters for poling, such as the strength of coercive and internal fields, and the relaxation time of the internal field, were not investigated, and an optimum poling condition was not established. Therefore, the period of the fabricated PPKN was not satisfactorily uniform. Consequently, the experimental effective nonlinear coefficient (3.7 pm/V) reached only 52% of the theoretical value, *d*_{eff}=(2/π)*d*₃₁.

In this letter, we report on the 180° domain switching characteristics and fabrication of very high quality PPKN for efficient second-harmonic generation. Potassium niobate (KNbO₃) belongs to the orthorhombic mm2 class with cell parameters *a* = 5.697, *b* = 3.971, *c* = 5.722 Å. Potassium niobate is ferroelectric at room temperature. As the temperature falls from the growth temperature (1333 K), it undergoes two phase transitions; one is from paraelectric (cubic) to ferroelectric (tetragonal) at 708 K and the other is from ferroelectric (tetragonal) to ferroelectric (orthorhombic) at 498 K. KNbO₃ crystals were grown by the top seeded solution growth method with pulling at the authors' laboratory. Since as-grown KNbO₃ crystals consist of 60°, 90°, 120°, and 180° domains in the orthorhombic phase,¹⁰ they have to be made into a single domain by applying a dc electric field of 400–600 V/mm along the polar *c*-axis before beginning the process of periodic poling. Thin plate samples were cut from the single domain crystal and polished perpendicular to the *c*-axis. The thickness and area of the sample were 0.6 mm and 9×8 mm², respectively.

The coercive and internal fields of a KNbO₃ crystal were determined by measuring a hysteresis loop, which was observed by the voltage across a 20 μC standard capacitor connected with the sample in series, using an applied field of triangular pulses with a slope of 9.5 V/mm per ms and a period of 200 ms. To characterize the switching dynamics of domains, the polarization current was also measured by monitoring the voltage across a 1 MΩ resistor that replaced the standard capacitor. A high voltage amplifier (Trek 20/20b) and a function generator (Tektronix™ AFG 310) were used for supplying high power pulses of the designed shape, and an oscilloscope (Tektronix TDS 380) was used to measure the voltage. LiCl-riched electrolyte was used for electrodes, the diameter of which was 2.4 mm. After poling was complete, both *c*+ and *c*- faces were either etched with 24% hydrofluoric acid for 2–5 min., or mechanically polished with SiO₂ suspension fluid, in order to reveal domain structure.

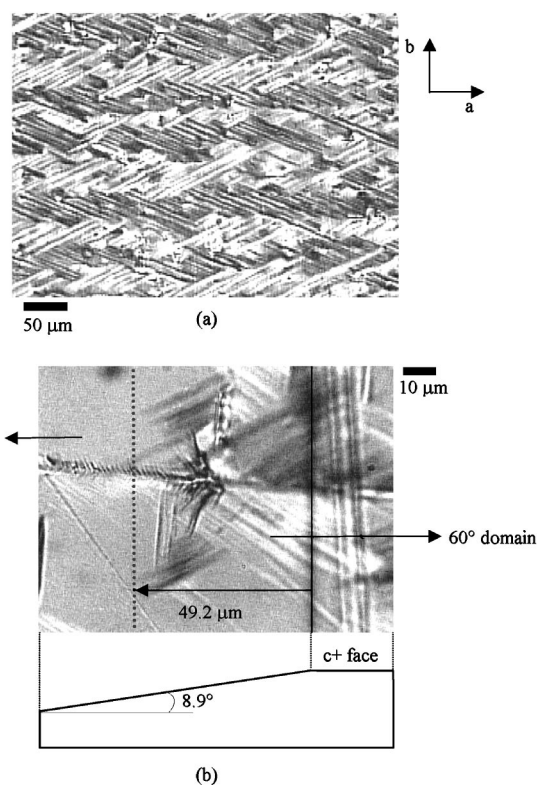
Figure 1 shows a hysteresis loop of the KNbO₃ crystal measured at room temperature. The spontaneous polarization

^{a)}Electronic mail: csyoon@mail.kaist.ac.kr

FIG. 1. A hysteresis loop of KNbO_3 .

is 0.42 C/m^2 , which is almost the same value as that reported previously.¹¹ The internal field E_{int} and coercive field E_c were 169 and 552 V/mm, respectively. The coercive field corresponds to the electric field required for domain wall motion. The internal field is caused by charge imbalances in the presence of lattice defects, such as ionic vacancies and antisites.¹² When polarization is reversed from one orientation to the opposite, the defect charges redistributed to minimize the total free energy of the ferroelectric system and a relaxation of internal field occurred over a period of time, which is called relaxation time. The relaxation times of the internal field in KNbO_3 for forward and reverse polings were 35.1 and 45.8 ms, respectively, and were measured by employing the method similar to the one used in LiNbO_3 .¹³ Two distinct relaxation times, fast (55.5 ms for forward poling and 125.2 ms for reverse poling) and slow (a few days), were reported in LiNbO_3 and a different type of defect is responsible for each relaxation time.¹² However, in KNbO_3 only fast relaxation was observed, suggesting that a single type of defect is responsible for the relaxation.

Four types of domain (60° , 90° , 120° , and 180° domains) were reported to exist in KNbO_3 crystals.¹⁰ When the poling field was below 600 V/mm, only 180° domains were observed. However, as the poling field rose higher than 600 V/mm, 60° domains were observed on the surface of a sample. A polarizing microscope image of the $c+$ face reveals that two types of 60° domains represent bright contrast and the other two types dark contrast [Fig. 2(a)]. To investigate how deep the 60° domains penetrate into volume, the $c+$ face was beveled with an angle of 8.9° . As shown in Fig. 2(b), the 60° domains disappear at $49.2 \mu\text{m}$ from the top edge, which is in the $c+$ face. Thus, the thickness of the 60° domains is estimated to be $49.2 \mu\text{m} \times \tan 8.9^\circ = 7.8 \mu\text{m}$. Once the surface layer was removed by polishing, the 180° domains remained intact, which confirms that the 60° domains exist only in the surface layer. The generation of the 60° domains in the surface layer in high electric fields may be ascribed to a neutralizing process of space charges that are placed in the surface layer of the 180° domains. In low electric fields, the speed of domain wall motion is slower than that of space charge redistribution. Therefore, the space charge relaxation provides a more effective way of stabilizing the 180° domains without resorting to the generation of 60° domains in the surface layer. However, as the applied field goes higher, the speed of domain wall motion exceeds

FIG. 2. Polarizing microscope images of (a) the $c+$ face with 60° domains and (b) the beveled $c+$ face.

the speed of space charge redistribution and the generation of the 60° domains in the surface layer is more convenient than the space charge relaxation.

Based on the findings above, periodically poled domain structures were fabricated using a single domain KNbO_3 crystal plate of $5 \times 9 \times 0.925 (a \times b \times c) \text{ mm}^3$. A periodic pattern of photoresist was formed on the $c+$ face using a standard photolithographic technique with a period of $32.5 \mu\text{m}$ and a duty ratio of 0.375. The period of $32.5 \mu\text{m}$ was obtained from the Sellmeier equations given in Ref. 14 for d_{31} ($= 11.9 \text{ pm/V}$) component. Although it was more advantageous to use d_{33} component, control of the QPM period for d_{33} at a wavelength of $1.064 \mu\text{m}$, which is $6.35 \mu\text{m}$, was beyond our fabrication capability. The region where photoresist was not deposited was in contact with liquid LiCl electrolyte and acted as an electrode. The entire $c-$ face was kept in contact with the liquid electrolyte. Since propagation velocity of the domain wall is much faster in the a -direction than in the b -direction, high quality periodic domain structures could be only obtained when the \mathbf{k} vector of the electrode structure was made to be parallel to the b -axis. The total area of poled region was $3 \times 5.3 (a \times b) \text{ mm}^2$, but the area of the periodically patterned electrode was $3 \times 4 (a \times b) \text{ mm}^2$. To avoid generating 60° domains in the surface layer and to facilitate the formation of 180° domains along the field direction, a modified triangular pulse was designed with a peak intensity of 525 V/mm, which consisted of a main part of triangular shape with a slope of $\pm 10.4 \text{ V/mm}$ per millisecond and a tail part with a slope of -1.1 V/mm per millisecond (Fig. 3, solid line). The function of the main part was to induce domain reversal and that of the tail part to prevent backswitching. Considering the relaxation time, a

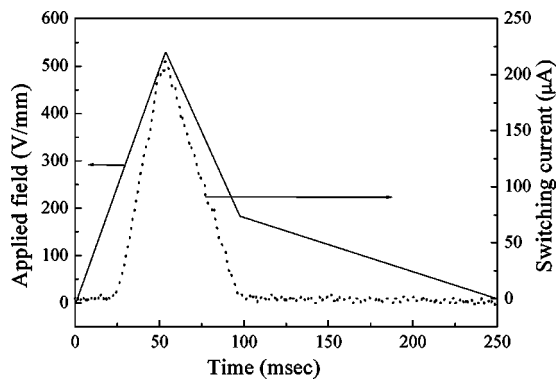


FIG. 3. Applied electric field pulse of a modified triangular shape (solid line) and the corresponding switching current (dotted curve).

single pulse with a duration of 250 ms, 100 ms for the main part plus 150 ms for the tail, was applied. The corresponding switching current is shown in Fig. 3 (dotted line). A total charge of $7.8 \mu\text{C}$ flowed to the poled region, which is 106% of the values calculated from the spontaneous polarization of 0.42 C/m^2 . Figure 4 shows a micrograph of the $c+$ and $c-$ faces of the periodically poled KNbO_3 crystal, which was polished with SiO_2 suspension fluid. The periodicity of the PPKN is well defined and uniform, indicating that the PPKN is of very high quality. The observed duty ratio of 0.41 is slightly larger than the duty ratio of the patterned electrode, 0.375, which indicates that a broadening of 9.3% of the 180° domain walls occurred during the poling process.

A 4-mm-long piece of PPKN with a $32.5 \mu\text{m}$ period was used for second-harmonic generation of a mode-locked Nd:YAG laser operated at 1064 nm with a pulse width of 35 ps and a repetition rate of 10 Hz. The fundamental beam was focused at a diameter of $300 \mu\text{m}$ and incident on the polished, uncoated b-face of the PPKN. The SHG pulse energy was measured with a power/energy meter (Scientech AC 2501) as a function of input pulse energy. Figure 5 shows that the SHG pulse energy increases nearly quadratically with the fundamental pulse energy and the maximum conversion efficiency of 34.8% was obtained at the fundamental

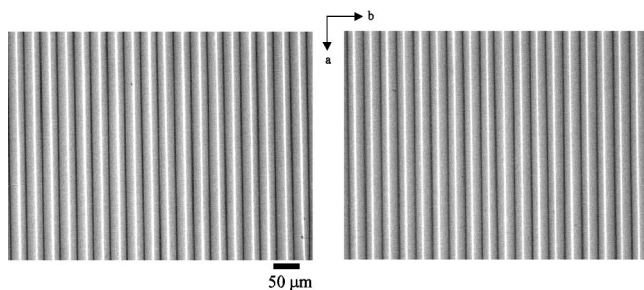


FIG. 4. $c+$ face (left) and $c-$ face (right) of PPKN with a domain period of $32.5 \mu\text{m}$.

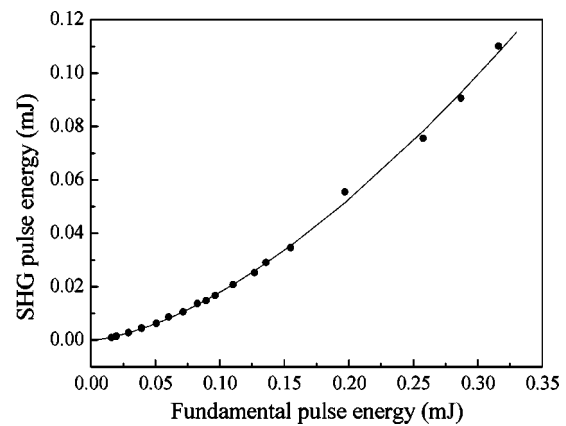


FIG. 5. SHG pulse energy as a function of fundamental pulse energy at 1064 nm wavelength for a 4 mm PPKN.

pulse energy of 0.32 mJ. The effective nonlinear coefficient was measured in the limit of undepleted pump approximation, using a 9 ns pulse width Nd:YAG laser to avoid group velocity mismatch, and $d_{\text{eff}}=5.2 \text{ pm/V}$ was obtained, which is 68% of the theoretical value, $(2/\pi)d_{31}$.

In summary, we investigated the domain switching characteristics of KNbO_3 and designed a pulse form of modified triangular shape based on the poling parameters determined from the investigation. By establishing optimum poling conditions, we fabricated periodically poled potassium niobate crystals with a dimension of $3 \times 4 \times 0.925 (a \times b \times c) \text{ mm}^3$ for quasi-phase-matched second-harmonic generation. The excellent quality of the fabricated PPKN was proved by the uniform domain pattern, a large effective nonlinear coefficient of $d_{\text{eff}}=5.2 \text{ pm/V}$ and a high SHG conversion efficiency of 34.8%.

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- ¹J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. Pershan, *Phys. Rev.* **127**, 1918 (1962).
- ²M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, *IEEE J. Quantum Electron.* **28**, 2631 (1992).
- ³L. E. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, W. R. Bosenberg, and J. W. Pierce, *J. Opt. Soc. Am. B* **12**, 2102 (1995).
- ⁴K. Mizuuchi and K. Yamamoto, *Appl. Phys. Lett.* **66**, 2943 (1995).
- ⁵G. Rosenman, A. Skliar, D. Eger, M. Oron, and M. Katz, *Appl. Phys. Lett.* **73**, 3650 (1998).
- ⁶H. Karlsson and F. Laurell, *Appl. Phys. Lett.* **71**, 3474 (1997).
- ⁷W. P. Risk and G. M. Loiacono, *Appl. Phys. Lett.* **69**, 311 (1996).
- ⁸Y. Yang and C. S. Yoon, *Appl. Phys. Lett.* **75**, 1164 (1999).
- ⁹J.-P. Meyn, M. E. Klein, D. Woll, and R. Wallenstein, *Opt. Lett.* **24**, 1154 (1999).
- ¹⁰*Modern Crystallography IV*, Springer Series in Solid State Science Vol. 15, edited by L. A. Shuvalov (Springer, Berlin, 1988), p. 209.
- ¹¹P. Gunter, *J. Appl. Phys.* **48**, 3475 (1977).
- ¹²V. Gopalan, T. E. Mitchell, Y. Furukawa, and K. Kitamura, *Appl. Phys. Lett.* **72**, 1981 (1998).
- ¹³J. H. Ro and M. Cha, *Appl. Phys. Lett.* **77**, 2391, (2000).
- ¹⁴B. Zysset, I. Biaggio, and P. Gunter, *J. Opt. Soc. Am. B* **9**, 380 (1992).