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ABSTRACT

A way to stabilize the domain structure in periodically poled Rb-doped $KTiOPO_4$ samples at high-temperatures is presented. The domain contraction along the *b*-crystallographic axis that is observed when crystals are annealed at high temperatures is suppressed when the ends of the domains along the *b*-axis are diced away. Additionally, the thermal stability of self-assembled domain gratings with a sub-μm average periodicity of 650 ± 200 nm and a domain-width of 225 ± 75 nm in mm-thick samples is investigated, and it is shown that the key factor for the domain stability is the domain width rather than the interdomain distance.

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Today it is possible to tailor many nonlinear optical processes by exploiting the quasi-phase matching (QPM) technique using domain-engineered ferroelectric crystals fabricated by electric field poling.[1,2] To obtain an efficient nonlinear response, the domain grating along the propagation direction has to be long and homogeneous and follow the designed pattern with high accuracy. The required domain period in the light propagation direction is on the order of a few micrometers, or shorter, and in the transverse dimensions, it has to be in the range of millimeters. Therefore, ferroelectric domains form thin sheets with high aspect ratios (width to depth), which means a large domain wall area to domain volume ratio. To date, periodically poled (PP) crystals from the $KTiOPO_4$ (KTP) family have been among the most commonly exploited for nonlinear optics, thanks to good power handling capability[3] and the possibility to obtain large apertures.[4] Rb-doped KTP (RKTP), in particular, has shown to be an exceptional material for the fabrication of sub-μm periodicities over mm-thick crystals[5,6] and has been exploited to fabricate high aspect-ratio domain gratings oriented along the *a*-direction, which has several important technical applications.[7–9]

We have recently shown that 1-mm-thick c-cut periodically poled RKTP (PPRKTP) samples show thermally induced anisotropic domain wall (DW) motion along the *a*- and *b*-crystallographic axes when they are annealed above 550 °C.[10] We found that bulk domain contraction along the *b*-axis was on the order of tens of micrometers, whereas in the *a*-direction it resulted in approximately three orders of magnitude lower domain wall displacement. However, domains that did not extend over the whole sample thickness (in the polar direction) merged or back-switched, with their motion along the a-direction. Furthermore, we found that the domain contraction strongly depends on the domain grating periodicity,[10] being considerably pronounced for periodicities below 9 μm. For applications where the samples are processed or treated at elevated temperatures, such as epitaxial growth using a periodically poled crystal as a seed,[11] or fabrication of waveguides[12] and other integrated devices, it becomes critical to be able to control the domain stability, especially when a short periodicity is required.[13] Moreover, it is important from a fundamental point of view to understand which of the two factors, i.e., the domain width or the grating periodicity, contributes to thermally induced domain instability.

In this work, we demonstrate that for RKTP the domain contraction along the *b*-direction, that is observed when the crystals are annealed at high temperatures, can be suppressed if

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the edges of the domains are diced away along the *a*-direction. We show that this method is effective for all the studied grating period, ranging from sub-μm to 9 μm periodicities. In addition, we show that the domain width affects the thermal instability rather than the inter-domain distance in short period samples.

In our experiments, we first studied 1-mm-thick *c*-cut RKTP samples exhibiting different domain grating periodicities: 9 μm and 3.18 μm. These samples were obtained using standard lithography and electric field poling.[14] Additionally, we investigated self-assembled domain gratings with sub-μm periodicities.[15] After electric field poling, all the samples were selectively etched in a KOH-KNO_3 solution at 100 °C in order to reveal the domain structure.[16] Some of the samples were then cut with a dicing saw along the *a*-direction, cutting away 0.3 mm of the domain grating, in order to terminate the *b*-face of the domain grating in air, as shown in Fig. 1(b). The cut extended throughout the whole sample length and thickness. From here on, we will refer to these crystals as "edge-cut" samples. All the crystals were then annealed in air for several hours at high temperatures. In all cases, the temperature was ramped up with a constant heating rate of 25 °C/min, and the cool-down rate was 2.4 °C/min. The samples were then selectively etched again in order to reveal any changes in their domain structure. Since each etching step creates a surface relief, it allowed us to study both the original and post-thermal-treatment domain configurations. Throughout this work, we refer to sample thickness as the dimension of the crystal along the polar c-direction and to domain width and domain length to the domain dimensions along the a- and b-directions, respectively.

Figure 1 shows optical micrographs of two PPRKTP samples with 9-μm-period domain gratings annealed at 730 °C for 12 h. Here, the domains extended through the whole thickness, and therefore only one of the polar faces is shown. Sample A (a) was non-edge-cut, while sample B (b) was edge-cut. For the non-cut sample, large domain contractions in the *b*-direction are clearly observed as an etched-step: the arrow in Fig. 1(a) shows the pre-annealing position of the domain wall. Note that the original domain configuration appears somewhat dimmer, the post-annealing one present a stronger contrast. The average domain-contraction along the *b*-axis is 59 μm for this sample, in good agreement with our previous results.[10]

On the other hand, no domain contraction is observed when the crystal has been edge-cut prior to annealing, as it is evident from Fig. 1(b). It has previously been suggested that during heat treatment, an internal field, large enough to induce domain wall (DW) motion, is created by the redistribution of bulk charges, ions and/or reorientation of defect dipoles.[11] The large DW movement along the *b*-axis compared to that along the *a*-axis can be attributed to the fact that the DW velocity in the *b*-direction is more than 30 times faster than along the a-axis.[17] On the other hand, if the domain ends are cut away, leaving their b-faces exposed to air, the DWs get pinned at the crystal edges, and the domain reversal is inhibited. Similar behavior has also been observed in stoichiometric lithium tantalate (SLT), where the domain stability could be largely increased by cutting the edges of periodically poled samples,[18] in a similar manner as we have done here, thereby eliminating the stored surface electrostatic energy of the domain ends.

The same experiment was performed with 3.18 μm-period PPRKTP samples. In this case, they were heat-treated at 730 °C for 4.5 h. For the non-cut samples, the contraction was measured to be on the order of 300–400 μm. As is expected,[10] this was considerably larger, about 7-times, than for the 9 μm-period samples. It can be seen in Fig. 2 that the edge-cutting method also effectively eliminated the contraction along the b-direction for these short-periodicity samples. Nevertheless, it is worth noting that some domains, those marked by the arrows in Fig. 2, have switched back, i.e., they have returned to the original polarization orientation they had prior to poling (clearly seen in the magnified picture in the inset). This back-switching was only observed for domains that had not propagated through the whole crystal bulk during periodic poling, and hence presented charged domain walls that were in a tail-to-tail (TT) configuration, which is energetically costlier. Thus, in order to minimize their energy, they back-switch at high temperatures. This back-switching is not prevented by the edge-cutting method, which

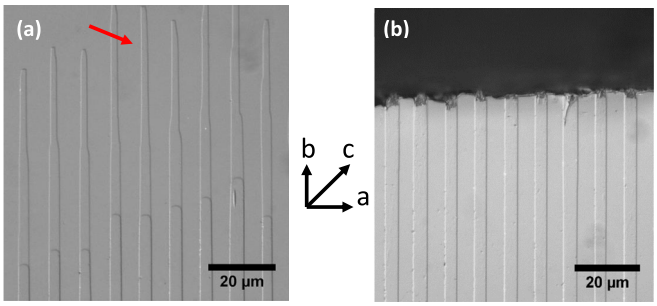

FIG. 1. Domain grating revealed by chemical etching. (a) A non-cut 9-μm period PPRKTP etched prior to and after heat treatment at 730 °C for 12 h. The arrow marks the pre-annealing position of the domain wall. The domains had shrunk ∼59 μm in the b-direction after heat treatment. (b) An edge-cut 9-μm-period RKTP where no contraction is observed.

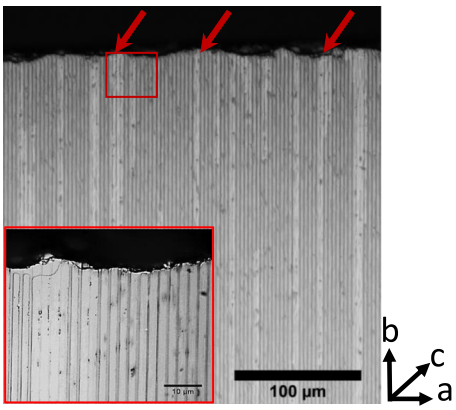

FIG. 2. Edge-cut 3.18-μm period PPRKTP etched prior to and after heat treatment at 730 °C for 4.5 h. Arrows show the TT domains which switched back during annealing. The inset shows a magnified picture of the square-marked region.

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indicates that the energy penalty for having TT domains cannot be compensated for by terminating their *b*-face to air.

Finally, we tested the edge-cutting method on RKTP samples with self-assembled sub-μm domain gratings (SA-RKTP).[15] SA-RKTP samples were fabricated by first submerging 1-mm-thick, single domain *c*-cut samples in a KOH/KNO_3 water-based solution with the molar ratio $KOH:KNO_3:H_2O$ of 1.8:1:11.2 at 135 °C for 3 min. Afterward, the samples were brought into contact with the poling circuit using planar liquid KCl electrodes and poled by applying single 5-ms long symmetric triangular E-field pulses of magnitudes ranging from 4.8 to 6.4 kV/mm. This method results in bulk quasi-periodic sub-μm domain gratings, consisting of thin strip domains oriented in the *b*-direction with an average domain periodicity of 650 ± 200 nm and a domain-width of 225 ± 75 nm.[15] It should be pointed out that these domains also extend through the whole crystal thickness. Note that depending on the magnitude of the applied field, SA-RKTP can be obtained with different domain densities, different inter-domain distances, and different domain widths. Previously,[10] we had observed complete back-switching during thermal annealing at 730 °C for such samples, which is ascribed to the fact that the domain contraction along the *b*-direction increases strongly with decreasing period,[10] eventually forming a single-domain state. Here, the edge-cut SA-PPRKTP as well as regular SA-PPRKTP, were annealed at 650 °C for 4.5 h. The regular SA-PPRKTP back-switched completely, while the edge-cut SA-PPRKTP presented similar results to those obtained with the 3-μm-period PPRKTP samples: the domains that extend through the whole sample thickness and present uncharged DWs parallel to the *c*-axis were well stabilized, but the domains that were in head-to-head (HH) or TT resulted in domain-merging or back-switching, as it was in good agreement with the results obtained in Ref. 10. Thus, although edge-cutting does not prevent back-switching for the HH or TT domain configurations, it is a powerful technique to prevent domain contraction along the *b*-direction, even for sub-μm periodicity samples.

Finally, we investigated the increase in domain contraction when the periodicity of the domain grating decreased,[10] in order to understand whether it is the domain width or the inter domain distance that is the determining factor for the domain contraction. Note that since in PPRKTP the domain width also changes with the periodicity, it is not possible to discern which of the two parameters is the critical factor by solely studying these samples. Therefore, we extended our study to SA-PPRKTP where both have different domain widths, and domain inter-distance are represented, making these crystals an ideal case-study to answer this question. Furthermore, note that all the domains used in this study had the same thickness of 1 mm.

Several SA-RKTP samples without edge-cutting were annealed at 350 °C for 1 h, and the amount of contraction along the *b*-axis was measured. Figure 3 shows the dependence of the contraction on the domain width (a) and the domain inter-distance (inset). It is evident that the observed contraction had an exponential dependence on the domain width, while the correlation with the inter domain distance is much less pronounced. Furthermore, Fig. 3(b), shows that for domains of the same width (700 nm in this case), the contraction is independent of the domain inter-distance.

Studies on domain stability in other materials like SLT or lithium niobate (LN) had reached different conclusions. Similar to this study, Liu *et al.* reported[19] that the stability of SLT domains decreased with decreasing domain size, but that the stability could be increased if the same domains were arranged in a grating. On the other hand, Kan *et al.*[20] studied congruent LN and found that the shorter the inter domain distance, the higher the domain instability. They ascribed their findings to the existence of a repulsive force between the DWs. For RKTP exhibiting domain widths of 225 nm and wider, we believe that the large internal screening, due to the mobile K^+ ions, can dramatically reduce the domain-domain interaction, and then the domain width becomes the critical factor for the domain stability. Narrow domains have a small domain volume to domain wall area ratio, making them more unstable.[21] Nevertheless, with the edge-cutting technique, in which the most energetically costly DWs, i.e., those cutting the a-axis, are taken out. This can be of utmost importance for optical applications requiring short-period PPRKTP devices, for which any post-poling treatment involving high temperatures will demand domain stabilization, and here the edge-cutting technique may play a crucial role.

In conclusion, we show that dicing the edge of domain gratings in RKTP along the *a*-direction in such a way that the *b*-faces of the periodic domain structure terminate in air improves the stability of the domain structure largely. This

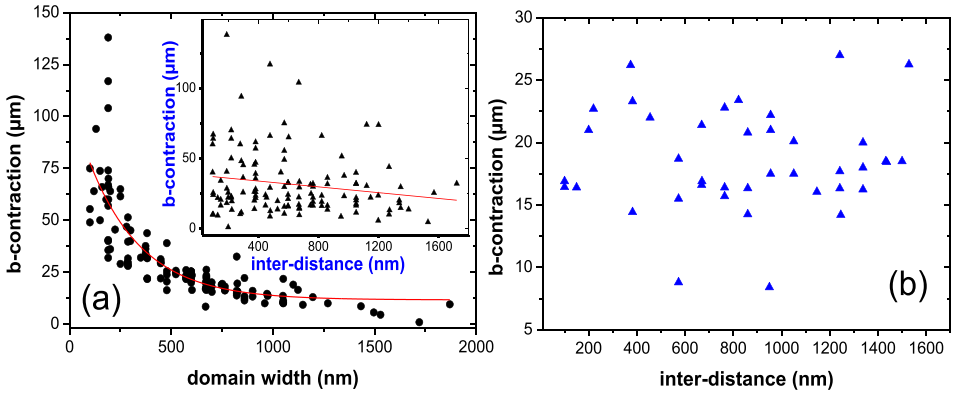

FIG. 3. (a) Domain contraction along the *b*-axis as a function of domain width after heat treatment at 350 °C during 1 h of annealing. An exponential dependence can be seen in the graph. The inset shows the dependency of contraction on inter-distance for all range of domain widths in SA-RKTP. In (b) is shown the dependence between domain contraction along the *b*-axis and the domain inter-distance for ∼700 nm domain width.

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method effectively works for grating periodicities ranging from several hundreds of nanometers to those that are of several micrometers wide. Moreover, SA-PPRKTP samples with varying domain width and domain inter-distance have been used to demonstrate that it is the domain width rather than the domain periodicity that is the main factor for increased instability in short period samples.

REFERENCES

[1]M. Yamada, N. Nada, M. Saitoh, and K. Watanabe, Appl. Phys. Lett. **62**, 435 (1993).
[2]J. D. Bierlein and F. Ahmed, Appl. Phys. Lett. **51**, 1322 (1987).
[3]J. Hellström, V. Pasiskevicius, H. Karlsson, and F. Laurell, Opt. Lett. **25**, 174 (2000).
[4]A. Zukauskas, N. Thilmann, V. Pasiskevicius, F. Laurell, and C. Canalias, Opt. Mater. Express **1**, 201 (2011).
[5]A. Zukauskas, G. Strömqvist, V. Pasiskevicius, F. Laurell, M. Fokine, and C. Canalias, Opt. Mater. Express **1**, 1319 (2011).
[6]C. Canalias, V. Pasiskevicius, R. Clemens, and F. Laurell, Appl. Phys. Lett. **82**, 4233 (2003).
[7]C. Canalias and V. Pasiskevicius, Nat. Photonics **1**, 459 (2007).
[8]C. Canalias, V. Pasiskevicius, M. Fokine, and F. Laurell, Appl. Phys. Lett. **86**, 181105 (2005).
[9]A. Zukauskas, A. L. Viotti, C. Liljestrand, V. Pasiskevicius, and C. Canalias, Sci. Rep. **7**, 8037 (2017).
[10]G. Lindgren, A. Peña, A. Zukauskas, C. Liljestrand, B. Ménaert, B. Boulanger, and C. Canalias, Appl. Phys. Lett. **107**, 082906 (2015).
[11]A. Peña, B. Ménaert, B. Boulanger, F. Laurell, C. Canalias, V. Pasiskevicius, P. Segonds, C. Félix, J. Debray, and S. Pairis, Opt. Mater. Express **1**, 185 (2011).
[12]M. G. Roelofs, P. A. Morris, and J. D. Bierlein, J. Appl. Phys. **70**, 720 (1991).
[13]A. Zukauskas, V. Pasiskevicius, and C. Canalias, Opt. Express **21**, 1395 (2013).
[14]H. Karlsson, F. Laurell, and L. K. Cheng, Appl. Phys. Lett. **74**, 1519 (1999).
[15]A. Zukauskas, V. Pasiskevicius, and C. Canalias, Appl. Phys. Lett. **103**, 252905 (2013).
[16]F. Laurell, M. G. Roelofs, W. Bindloss, H. Hsiung, A. Suna, and J. D. Bierlein, J. Appl. Phys. **71**, 4664 (1992).
[17]C. Canalias, J. Hirohashi, V. Pasiskevicius, and F. Laurell, J. Appl. Phys. **97**, 124105 (2005).
[18]X. Liu, K. Kitamura, K. Terabe, and S. Takekawa, J. Appl. Phys. **102**, 014101 (2007).
[19]X. Liu, K. Kitamura, and K. Terabe, Appl. Phys. Lett. **89**, 142906 (2006).
[20]Y. Kan, H. Bo, X. Lu, T. Xu, Y. Jin, X. Wu, F. Huang, and J. Zhu, Appl. Phys. Lett. **97**, 202903 (2010).
[21]M. Y. Gureev, P. Mokrý, A. K. Tagantsev, and N. Setter, Phys. Rev. B-Condens. Matter Mater. Phys. **86**, 104104 (2012).

25 August 2023 11:54:12

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