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Nonlocal erasing and writing of ferroelectric domains using a femtosecond laser in lithium niobate

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We experimentally demonstrate the highly-efficient nonlocal erasing and writing of ferroelectric domains using a femtosecond laser in lithium niobate. Based on the induction of a focused infrared femtosecond laser without any relative displacement or additional treatment, the original multiple ferroelectric domains can be either erased (erasing operation) or elongated (writing operation) simultaneously in the crystal, depending on the laser focusing depth and the laser pulse energy. In the erasing operation, the original multiple ferroelectric domains can be cleared completely by just one laser induction, while in the writing operation, the average length of the ferroelectric domains can be elongated up to 235 μm by three laser inductions. A model has been proposed in which a thermoelectric field and a space charge field are used cooperatively to successfully explain the mechanism of nonlocal erasing and writing. This method greatly improves the efficiency and flexibility of tailoring ferroelectric domain structures, paving the way to large-scale all-optical industrial production for nonlinear photonic crystals and nonvolatile ferroelectric domain wall memories. © 2024 Optica Publishing Group

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Ferroelectrics are materials that exhibit spontaneous polarization below the Curie temperature [1]. Tailoring domain structures in ferroelectrics have been used in various fields, such as nonlinear photonic crystals [2,3], nonvolatile ferroelectric domain wall memories [4], domain wall electronics [5], and quantum optics [6,7]. The techniques to tailor ferroelectric domain structures include writing domains and erasing domains. The main methods for writing ferroelectric domains include electric field poling [8], all-optical poling [9], light-assisted electric field poling [10], probe-tip poling [11], and electron-beam poling [12]. Electric field poling is the most common approach, which inverts domains by applying an external electric field along the polar z axis of the crystal. Electric field poling, however, turns from tedious processing steps, and it is hard to handle thick crystals and fabricate domains with high precision [13].

All-optical poling is a highly promising approach due to its nonlocal manipulation, high precision, and the ability of 3D structure processing [14]. All-optical poling mainly includes UV light poling and focused infrared femtosecond laser poling. UV light poling can invert domains by the pyroelectric or

thermoelectric field, but the domain inversion is restricted to a shallow surface layer (several hundred nanometers) due to the strong absorption of UV light [15]. Focused infrared femtosecond laser poling can invert domains inside the crystal, but the domain length only reaches 60 μm [16]. In addition, the fabrication speed of domains is restricted to 10 $\mu\text{m}/\text{s}$ as domains are only inverted at the focal point.

To improve the fabrication efficiency, two-step laser poling has been proposed by us [17]. The first step is laser marking, in which a laser is used to mark the origin of the desired domain inversion. The second step is laser induction, in which a laser-induced thermoelectric field is used to invert the domain at the marker site and drive the domain growth. Two-step laser poling can manipulate the domain inversion and growth length of multiple marked sites within a diameter of $\sim 200 \mu\text{m}$ simultaneously, but the maximum domain length only reaches 130 μm . The fabrication efficiency can also be improved by the laser-driven strategy [18] and the pyroelectric field-assisted strategy [19,20].

The main methods for erasing ferroelectric domains include femtosecond laser erasing and probe-tip erasing. Femtosecond laser erasing can selectively erase the previously written domain by moving the laser along the $+z$ direction, fabricating nanodomains beyond the diffraction limit [21]. Still, the erasing speed of domains is restricted to 10 $\mu\text{m}/\text{s}$ as domains are only erased at the focal point. Probe-tip erasing uses PFM to erase domains on the nanoscale by applying a D.C. voltage to the probe tip and has been used to fabricate nonvolatile ferroelectric domain wall memories. However, it is only capable of erasing domains at the surface of crystals [22,23].

In this paper, we experimentally demonstrate a new nonlocal erasing and writing approach for ferroelectric domain engineering in lithium niobate. It is based on the induction of a focused infrared femtosecond laser without any relative displacement or additional treatment. The laser induction has two operations: the erasing operation and the writing operation, depending on the laser focusing depth. The original multiple domains within a diameter of $\sim 200 \mu\text{m}$ can be either cleared completely in the erasing operation or elongated simultaneously in the writing operation. Additionally, after being cleared completely, the original domain structures can be written again by a single writing operation. This technique greatly improves the efficiency and flexibility of tailoring ferroelectric domain structures and is also suitable for other ferroelectrics.

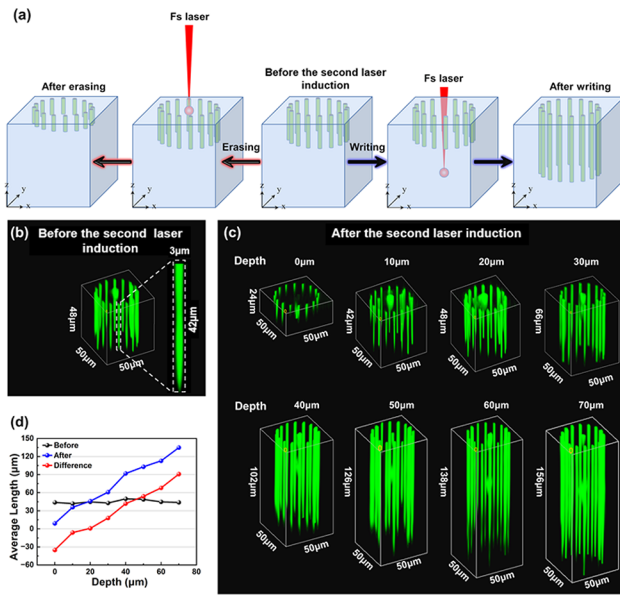


Fig. 1. (a) Experimental scheme. The second laser induction is the erasing operation when the laser focusing depth is less than a depth D and is the writing operation when the laser focusing depth is more than D . (b) Original domains before the second laser induction. (c) Results after the second laser induction with different laser focusing depths. (d) Difference in the average length of domains before and after the second laser induction with different laser focusing depths. As the randomness of the “quality” of the markers [17], there will be fluctuations in the average length of the original domains fabricated by the same parameters. The domain structures are visualized by Čerenkov-type SH confocal microscopy.

In the experiment, we use 500- μm -thick z -cut 5% MgO -doped LiNbO_3 crystals. The LiNbO_3 sample is mounted on a three-axis XYZ displacement stage (Aerotech Inc.) with a precision of 100 nm. The light source is output from a regenerative amplified Yb:KGd(WO₄)₂ (KGW)-based laser (Pharos, Light Conversion), which works at a wavelength of 1026 nm, a pulse duration of 190 fs, and a repetition rate of 1000 kHz. The pulse energy can be continuously adjusted from 0 to 400 μJ . The laser is focused by a microscope objective ($\times 50$, $\text{NA} = 0.42$), creating a focal spot diameter of about 1–2 μm . The domain structures are visualized by Čerenkov-type second harmonics (SH) confocal microscopy (FVMPE-RS, Olympus), allowing for nondestructive characterization [24,25].

The experimental scheme of nonlocal erasing and writing of ferroelectric domains using a femtosecond laser is shown in Fig. 1(a). A femtosecond laser is focused near the original domains without any relative displacement or additional treatment. Induced by the laser with different focusing depths, all of the original domains can be either erased (erasing operation) or elongated (writing operation) simultaneously. We experimentally researched the effect of the laser focusing depth on the original domains. As shown in Fig. 1(b), the original domain structure with an average domain length of $\sim 45 \mu\text{m}$ was fabricated by two-step laser poling [17], in which a marker ring with a diameter of 40 μm was fabricated on the crystal surface by the laser marker with a pulse number of 1 and pulse energy of 150 nJ, and the first laser induction was performed in the center of the ring of 20 μm below the crystal surface using a laser with a pulse number of 500,000 and pulse energy of 1100 nJ. Then

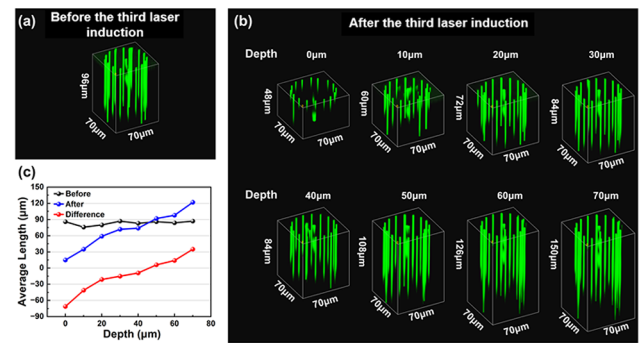


Fig. 2. (a) Original domains before the third laser induction. (b) Results after the third laser induction with different laser focusing depths. (c) Difference in the average length of domains before and after the third laser induction with different laser focusing depths. The domain structures are visualized by Čerenkov-type SH confocal microscopy.

the second laser induction was performed. A laser with a pulse number of 500,000 and pulse energies of 700, 900, 1100, 1300, 1500, 1700, 1900, and 2100 nJ was focused at the center of the marker ring of 0, 10, 20, 30, 40, 50, 60, and 70 μm below the $+z$ -surface of the crystal, respectively. The experimental results are shown in Figs. 1(c) and 1(d).

As shown in Figs. 1(c) and 1(d), domains were either erased or elongated after the second laser induction, closely related to the laser focusing depth. When the laser focusing depth was less than 20 μm , domains were erased. When the laser focusing depth was equal to 20 μm , the domain length was unchanged basically. When the laser focusing depth was more than 20 μm , domains were elongated, and the deeper the laser focusing depth was, the larger the increase in the domain length was. Additionally, laser induction led to permanent defects in the focal spot.

To further investigate the relationship between the laser focusing depth and the erasing and writing of domains, the original domains with an average length of $\sim 85 \mu\text{m}$ and a marker ring diameter of 60 μm were fabricated by the laser marker and two laser inductions, as shown in Fig. 2(a). The parameters of the laser marker and the first laser induction were the same as in Fig. 1(b), and the depth of the second laser induction with a pulse energy of 1500 nJ was 40 μm . Similarly, the original domains were induced by the third laser induction with different depths, using the same parameters as in Fig. 1(c). The experimental results are shown in Figs. 2(b) and 2(c). Domains were erased when the laser focusing depth was less than 40 μm and elongated when the laser focusing depth was more than 50 μm . Assuming the domain length is L , we can find that the second or third laser induction is the erasing operation when the laser focusing depth is less than $L/2$ and is the writing operation when the laser focusing depth is more than $L/2$. Notably, if the distance from the domain to the laser spot is less than a certain distance, which is about 10 μm here, the result will be different. In this case, the domain will be erased when the laser focusing depth is less than L and elongated when the laser focusing depth is more than L .

In addition to the laser focusing depth affecting the domain length, the laser pulse energy also affects it. Therefore, we experimentally researched the effect of pulse energy on domain length in erasing and writing operations. The original domains before the second laser induction were the same as in Fig. 1(b). In the

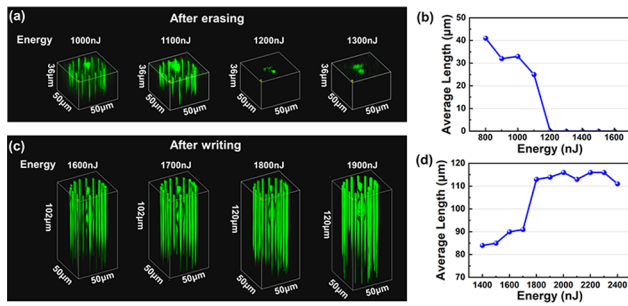


Fig. 3. (a) Results after erasing with different pulse energies. (b) Average domain length after erasing with different pulse energies. (c) Results after writing with different pulse energies. (d) Average domain length after writing with different pulse energies. The domain structures are visualized by Čerenkov-type SH confocal microscopy.

erasing operation, a laser with a pulse number of 500,000 and pulse energies of 800–1600 nJ was focused at the center of the marker ring of 10 μm below the +z-surface. The experimental results are shown in Figs. 3(a) and 3(b). The average domain length decreased with the increase of pulse energy. When the pulse energy exceeded 1200 nJ, the domain length decreased to 0, and domains were cleared completely. In the writing operation, a laser with a pulse number of 500,000 and pulse energies of 1400–2400 nJ was focused at the center of the marker ring of 50 μm below the +z-surface. The experimental results are shown in Figs. 3(c) and 3(d). The average domain length gradually increased with the increase of pulse energy at the beginning and finally reached a certain length and no longer increased. Therefore, domains can be erased or elongated to a specific length by adjusting the laser focusing depth and the laser pulse energy synergistically.

Notably, as the increase of the distance from the domain to the laser focal spot will increase the difficulty of elongating the domain, domain growth can be driven continuously through multiple laser inductions. As shown in Fig. 4(a), the original domains in Fig. 1(b) were elongated continuously by three writing operations, in which the laser focusing depths were 60, 100, and 140 μm, respectively. The length of domains generally exceeds 230 μm, and the average length is elongated up to 235 μm. Moreover, the longest domain is elongated up to 246 μm, which is far beyond the capability of the focused infrared femtosecond laser poling (60 μm) [16], two-step laser poling (130 μm) [17], and laser-driven domain growth (155 μm) [18]. Further optimizing the experimental parameters can fabricate longer domains.

Additionally, the combination of erasing and writing operations allows for more flexible and highly efficient fabrication of domain structures. As shown in Fig. 4(b), firstly, the tailored domain structures were cleared completely by an erasing operation. Then the original domain structures can be written again by a writing operation as the marker sites were still present. As shown in Fig. 4(c), at first, only a few of the domains were erased by adjusting the laser pulse energy and the laser focusing position in the erasing operation, and then the erased domains were written again by a writing operation. This implies that the nonlocal erasing and writing of ferroelectric domains not only can be used for the fabrication of nonlinear photonic crystals but also have potential applications in nonvolatile ferroelectric

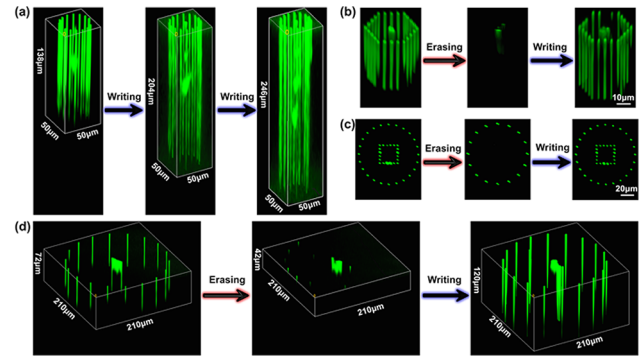


Fig. 4. (a) Average length of domains was elongated up to 235 μm after three writing operations. (b) Cleared the tailored domain structures completely by an erasing operation with a depth of 10 μm and pulse energy of 1300 nJ and wrote the original domain structures again by a writing operation with a depth of 30 μm and pulse energy of 1300 nJ. (c) Erased a few of the domains by an erasing operation with a depth of 10 μm and pulse energy of 1000 nJ and wrote the erased domains again by a writing operation with a depth of 20 μm and pulse energy of 1100 nJ. The image is in a depth of 5 μm. (d) Erased domains within a diameter of 200 μm simultaneously by an erasing operation with a depth of 10 μm and written domains again by a writing operation with a depth of 30 μm. The domain structures are visualized by Čerenkov-type SH confocal microscopy.

domain wall memories [26,27]. Moreover, erasing and writing operations have a wide manipulation range. As shown in Fig. 4(d), a laser induction with a pulse energy of 2000 nJ can erase or write multiple domains within a diameter range of 200 μm simultaneously. This implies that the efficiency of all-optical tailoring domain structures can be improved by several orders of magnitude.

The experimental results prove that multiple ferroelectric domains can be erased or elongated simultaneously by the induction of a focused infrared femtosecond laser without any relative displacement or additional treatment. To explain this effect, we propose a new model comprised of a thermoelectric field and a space charge field. The laser is tightly focused inside the crystal, leading to a local light heating process. The local heating process leads to a bipolar thermoelectric field (owing to the temperature gradient) and a pyroelectric field (owing to the time-varying temperature) [21,28]. Additionally, the increased temperature will reduce the coercive field for domain inversion. Previously, we have used the optically induced 3D thermoelectric field to explain the induction of domains [17], but it cannot explain the results. Because if the erasing and writing of ferroelectric domains are driven only by the thermoelectric field, domains will be erased when the laser focusing depth is less than the domain length L and elongated when the depth is more than L . Additionally, as the pyroelectric field did not induce domain inversion again at the marker sites after completely erasing domains, it is about several orders of magnitude smaller than the coercive field near the domains. Therefore, we can neglect the effects of the pyroelectric field.

As shown in Fig. 5(a), the thermoelectric field E_t can be divided into two parts: E_{t1} and E_{t2} . Due to its high conductivity [22,26,29], the ferroelectric domain wall will trap bulk and surface charges under the effect of E_t , resulting in a space charge field E_{sc} [19,20]. The thermoelectric field E_t and the space charge field E_{sc} together form the electric field E . When the distance

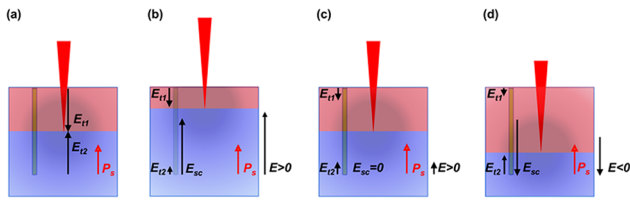


Fig. 5. (a) Laser induces a bipolar thermoelectric field inside the crystal. (b) E_{sc} is parallel to E_{t2} when the laser focusing depth is less than $L/2$. (c) E_{sc} is close to 0 when the laser focusing depth is equal to $L/2$. (d) E_{sc} is parallel to E_{t1} when the laser focusing depth is more than $L/2$.

from the domain to the laser spot is less than $10\ \mu\text{m}$, the thermoelectric field E_t plays a dominant role in the erasing and writing of the domain. However, as the distance is more than $10\ \mu\text{m}$ in the experiment, the strength of E_t near the domains decreases sharply with the increase of the distance. Additionally, the trapped charges gather on the domain walls, so E_{sc} is much stronger than E_t near the domain, playing a dominant role in the erasing and writing of the domain.

As shown in Figs. 5(b)–5(d), we assume that E_{t1} and E_{t2} induce a space charge field parallel to its direction on the domain walls at first. Finally, as the space charge field E_{sc} is unidirectional, it will be parallel to the thermoelectric field in which most of the domain wall is located. The larger the difference in the length of the domain in E_{t1} and E_{t2} , the larger the strength of E_{sc} . This explains why the domain is erased when the laser focusing depth is less than $L/2$ and elongated when the laser focusing depth is more than $L/2$. When the laser focusing depth is equal to $L/2$, E_{sc} will be close to 0, while the thermoelectric field E_{t2} near the domain is less than the coercive field. Therefore, the domain length will be unchanged basically.

Additionally, the laser pulse energy directly determines the strength of E_t , and the strength of E_t determines the concentration of charges, which consequently determines the strength of E_{sc} . Therefore, the domain length with the increase of the pulse energy will decrease in the erasing operation and increase in the writing operation. Notably, E_{sc} and E_{t1} will inhibit the erasing of domains when they are erased to a length of less than $10\ \mu\text{m}$ in Figs. 3(a) and 3(b). However, as the domain inversion is driven not only by the electric field but also by the energy of the displaced atoms after domain inversion [17], domains can be cleared completely when the pulse energy exceeds 1200 nJ. E_{sc} and E_{t2} will inhibit the writing of domains when they are elongated to a length of more than $100\ \mu\text{m}$ in Figs. 3(c) and 3(d), so the domain length reaches a certain length and no longer increases.

In conclusion, we have experimentally demonstrated the highly efficient nonlocal erasing and writing of ferroelectric domains using a focused infrared femtosecond laser without any relative displacement or additional treatment in lithium niobate. Using this method, two operations can be performed within a diameter of $\sim 200\ \mu\text{m}$: the erasing operation and the writing operation. The main difference is that the laser focusing depth is less than half of the domain length in the erasing operation, while it is more than half of the domain length in the writing operation. The original multiple ferroelectric domains can be cleared completely by an erasing operation, and the average length of the

ferroelectric domains can be elongated up to $235\ \mu\text{m}$ by three writing operations; all three operations take only 1.5 s. By properly designing the fabrication strategy, arbitrary 2D ferroelectric domain structures can be cleared completely and written again. A model has been proposed in which a thermoelectric field and a space charge field are used cooperatively to successfully explain the mechanism of nonlocal erasing and writing. The method provides a highly efficient and flexible tool for large-scale all-optical ferroelectric domain engineering, and it is also suitable for other ferroelectrics by acclimating the experimental parameters.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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