Ultrathin Slices of Ferroelectric Domain-Patterned Lithium Niobate by Crystal Ion Slicing

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ABSTRACT

We report the successful fabrication of 6 μm thick slices from a ferroelectric domain micro-engineered $LiNbO_3$ wafer device using the crystal ion slicing technique. The device was created by micropatterning ferroelectric domains in a bulk 0.3 mm thick wafer of z-cut $LiNbO_3$, followed by ion-implanting with 3.8 MeV He^+ ions to a fluence 5 x 10^{+16} ions/cm^2 to create a damage layer at a well defined depth from the surface. Etching away this damaged layer in dilute hydrofluoric acid results in a liftoff of the top slice in which the ferroelectric domain patterns are left intact. The influence of annealing conditions on liftoff time and depth of etch lines was studied. Helium-Neon laser light was successfully coupled into the device. Due to unintentional breakage of the polished input and output faces, the electro-optic scanning performance has not been characterized so far.

INTRODUCTION

The ability to control the angular position of a laser beam with high speed is of interest in many applications including optical communications, optical data storage, laser printing, and display technologies. Active solid-state electro-optic scanners based on micro-patterned $LiNbO_3$ have several advantages over mechanical and other systems including small device size and high operating speed (intrinsic response frequencies >100GHz). However, widespread application of these devices is currently limited because of the large voltage required to operate devices fabricated in single crystal wafers. A solution to this problem is to make the devices thinner, thereby reducing the operating voltage for a fixed driving electric field. However, the fabrication of high-quality thin films of $LiNbO_3$ has so far been elusive. Techniques such as pulsed laser deposition[1] and chemical vapor deposition[2] yield polycrystalline films, which have electro-optic responses much smaller than single crystal bulk $LiNbO_3$. Further, the light propagation losses in thin films are still high, which makes them unsuitable for optical devices.[3]

Recently, an ion-implantation-based technique called crystal ion slicing (CIS), has been reported for making thin slices of ferroelectric lithium niobate.[4,5] CIS allows a free standing micron-thick single crystal film to be fabricated from a bulk crystal. In CIS, ion implantation is used to define a damaged sacrificial layer several microns below the surface. This sacrificial layer is then etched away by immersion in a dilute hydrofluoric acid solution. The large etch selectivity of the damage layer over the rest of the crystal allows for the lifting off of a thin film, whose thickness is well controlled by the implantation energy.

By crystal ion slicing of domain-engineered devices fabricated on wafers, operating voltages can be significantly reduced. In this paper, we show that indeed domain-micropatterned

devices can be sliced into thin layers without destroying the domain patterns defined in the original crystal. We first describe the principle and fabrication of an electro-optic device used for laser beam scanning, followed by a description of the CIS processing of the fabricated device.

Electro-optic Scanner device on $LiNbO_3$

The most basic geometry for deflecting light is by use of an optical prism. In paraxial approximation, where the deflection angles are small, one can use a series of prisms in sequence, each prism successively deflecting the light beam further and further from the optical axis as shown in Fig. 1.

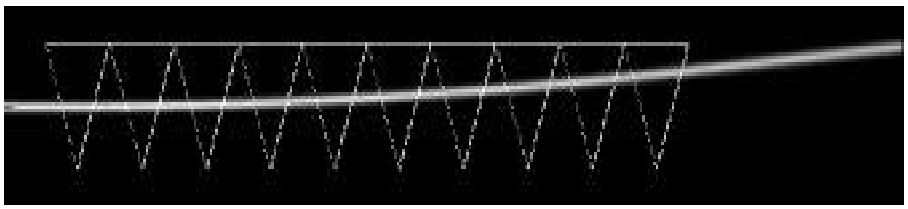

Figure 1: A beam propagation method (BPM) simulation of a rectangular scanner in $LiNbO_3$ operating at +15 kV/mm. A total of 10 triangles is shown, each triangle is 150 μm wide by 900 μm high. The total deflection is 4.3° from center, which gives 8.6° total deflection for ± 15 kV/mm.

The simplest scanner design is a rectangular scanner geometry which consists of N identical prisms placed in sequence, each with a base l and height W, such that the total length of the rectangular scanner is L=Nl, and width is W. The total deflection angle, θ_{int}, at the output for a light beam incident along the axis, L, of the rectangular scanner is given by[6]

$$\theta_{int} = \frac{\Delta n}{n}\frac{L}{W} \qquad (1)$$

where n is the index of the material, and Δn is the index difference between the prism and the surrounding matrix, where Δn<<n. These prisms are fabricated in an electro-optic medium where the refractive index n, and therefore the Δn is electric field tunable. This results in an electric field controlled deflection angle θ_{int}.

In a uniaxial electro-optic crystal such as $LiTaO_3$ or $LiNbO_3$, the refractive indices are electric field tunable. If the uniaxial direction is denoted subscript 3, then a decrease in the refractive extraordinary index n_e (or n_3) with the application of an electric field E_3 parallel to the spontaneous ferroelectric polarization direction is given by $\Delta n_e = -\frac{1}{2} n_e^3 r_{33} E_3$. If ferroelectric domains were created in the shape of a series of prisms in a z-cut crystal of $LiNbO_3$ or $LiTaO_3$, one can create an electro-optic scanner. Without an electric field, the refractive indices inside and outside the domain prisms will be equal, thus resulting in no deflection of light. When a uniform electric field is applied across the z-cut crystal in the z-direction, the electric field E_3 is parallel to the spontaneous polarization in one domain and antiparallel in the other. This results in an index difference in the extraordinary index of (n_{prism}-n_{matrix})= +/- 2Δn_e across the ferroelectric domain walls defining the prisms. Such rectangular scanners in $LiNbO_3$ and $LiTaO_3$ have been demonstrated.[7,8,9]

In $LiNbO_3$ the coercive field strength (the field required to reverse the domain at room temperature), E_c, is ~21 kV/mm.[10] So in a scanner as previously proposed, the maximum device

operating field strength should be less than the coercive field strength, so as to not destroy the domain patterns. On the other hand, the field should be as high as possible to achieve maximum deflection. For a typical field strength of 15 kV/m, if the device was fabricated in a commercially available crystal of thickness 0.3 mm, this would mean that the device would operate at (15 kV/mm)(0.3 mm) = 4500 V. This high operating voltage is a major hurdle to broader application of this technology, since electrical drivers that can provide large voltages at high bandwidths of GHz are not practical. A solution to this is to make the crystal thinner, so as to reduce the operating voltage.

EXPERIMENTAL DETAILS

A bulk device was fabricated on a 300 ± 5 μm thick, Z-cut single crystal, single-domain $LiNbO_3$ wafer. Domain micropatterning was performed as follows. A photoresist pattern resembling the device shown in Fig. 1 was defined on the +z face of the crystal surface with conventional contact photolithography. A uniform Tantalum (Ta) electrode was then deposited on the sample by DC sputtering. This was followed by liftoff process, which removes the metal in areas where photoresist exists beneath, by dissolving the underlying photoresist pattern in acetone. This leaves behind a complementary Ta-film pattern on the crystal surface resembling Fig. 1, with the Ta defining the complement of the area of the prisms composing the scanner. A photoresist coating is then deposited uniformly on the surface of the sample, across both the electrode and crystal features and baked well to remove any solvents. This layer has been found to improve selectivity of the domain reversal process by suppressing domain reversal beyond electrode edges through reduction of surface conduction in areas without the electrode. For domain inversion, ~21kV/mm electric field is applied across the crystal. The positive electrode is the Ta-film pattern on +z face while a uniform water electrode is used on the -z face as the ground. After domain reversal, the metal electrode is then removed by etching in HF.

This bulk device with patterned domain is then ion implanted with a dose of $5 \times 10^{+16}$ ions/cm^2 at an energy of 3.8 MeV with the temperature of the sample kept below 12°C during implantation. The input faces of the implanted devices are then end polished to 0.05 μm finish. After polishing, optimal condition rapid thermal annealing at 450°C for 40 s in flowing forming gas (5% H_2, 95% Ar) was used to enhance the etch selectivity between sacrificial layer and the rest of the sample. The device was then encapsulated in an HF resistant wax, which was used to protect the film's top surface and polished ends. Finally the device is immersed in dilute HF (10%) until it is completely detached. After detachment, electrodes were applied on each face with sputtered gold, and the sample (or sample fragments) were mounted and electrical contacts made to the surfaces. A beam of He-Ne laser light was focused to 2.5 μm in the vertical and 100 μm in the horizontal and targeted on the input faces of the device. The output face was imaged by a microscope objective and the light collected in a CCD camera.

DISCUSSION

The intermediate annealing step in the liftoff process above, allows relatively large sample areas (2 mm x 10 mm) to be lifted off in ~32 hours. This is believed to occur by promoting the diffusion of helium into the implantation induced micro voids and the buildup of an internal pressure in the sacrificial layer. Changing the annealing temperature and atmosphere had an effect upon both the time required for lift off and the quality of the lifted off slice. In

small test pieces (1.5 mm x 5 mm) of $LiNbO_3$ implanted at the same energy some general trends were noticed. In general, annealing temperatures lower than 450°C took longer to lift off as shown in Figure 2(a). For example, the lift-off times were 6 hours at 450°C, ~12 hours for 400°C, and >24 hours with no annealing. Annealing temperatures higher than 450°C tended to cause spalling. Other annealing atmospheres, such as Ar, N_2 and air, caused longer lift off times. The liftoff face showed etch lines that were characterized by atomic force microscopy as shown in Figure 3. The optimal lift conditions yielded an etch depth of 20.2 nm, which was much better than >40 nm for other processing conditions. The density of the etch lines was found to have no clear relation between process atmosphere, temperature, or exposure time to acid.

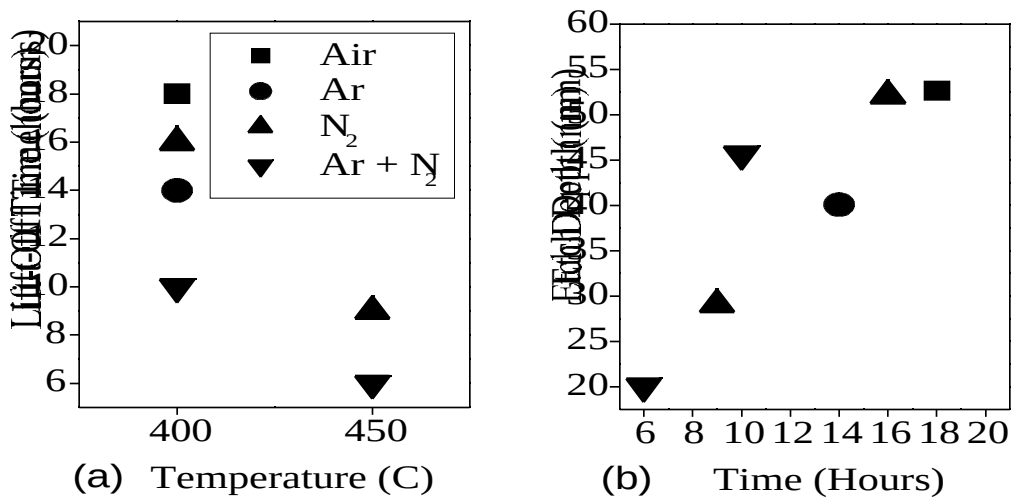

Figure 2: (a) Lift off time increased for higher processing temperatures. (b) Etch depth was found to scale linearly with exposure to acid

A CIS domain engineered device 2 mm x 9 mm was successfully lifted off using optimal conditions. Figure 4 shows a composite optical micrograph of the device. The domain structures patterned in the crystal before the implantation, annealing, and lift off step are still present in the crystal. The crystal is 6 μm thick, which was determined by imaging the crystal edge under a microscope. Figure 4 also shows some of the inherent difficulties in lifting off large size devices. Notice the large chip out of the input face and damage to the output face. If these chips are in the entrance or exit path for the scanner channel, no light could be coupled. Also, the lifted off scanners tended to show curvature in the plane. This curvature caused cracking and breaking of the sample in subsequent processing steps, like electrode coating and mounting. Because of these problems, only partial device fragments were mounted and tested in this study. Light from a He-Ne laser was successfully coupled into a fragment with a polished output face and an input face which was unintentionally fractured. Figure 5 shows the light exiting the output face, as imaged by a Charge Coupled Device(CCD) camera. However, since the light was not in a scanner channel in this region, the device scanning could not be tested in this work. However, electro-optic modulation has been previously reported in similar CIS sliced pieces, and we believe these devices would work with better input and output faces.[11]

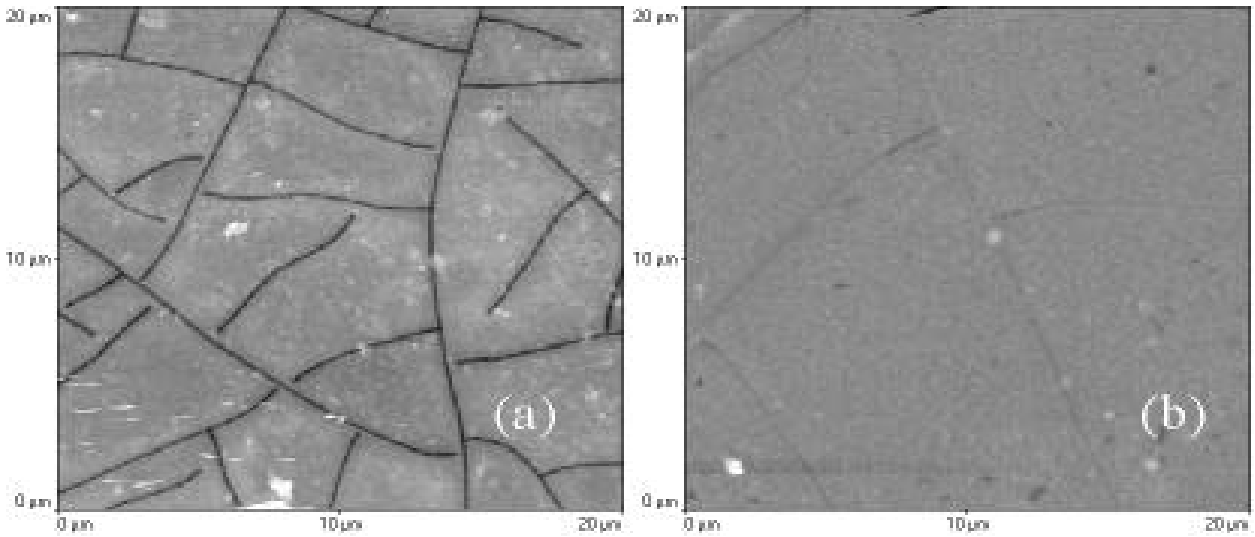

Figure 3. AFM images (a) 400°C in N_2 lift off for 16 hours with etch depth of 50.52 nm. (b) 450°C in forming gas lift off for 6 hours with etch depth of 20.31 nm.

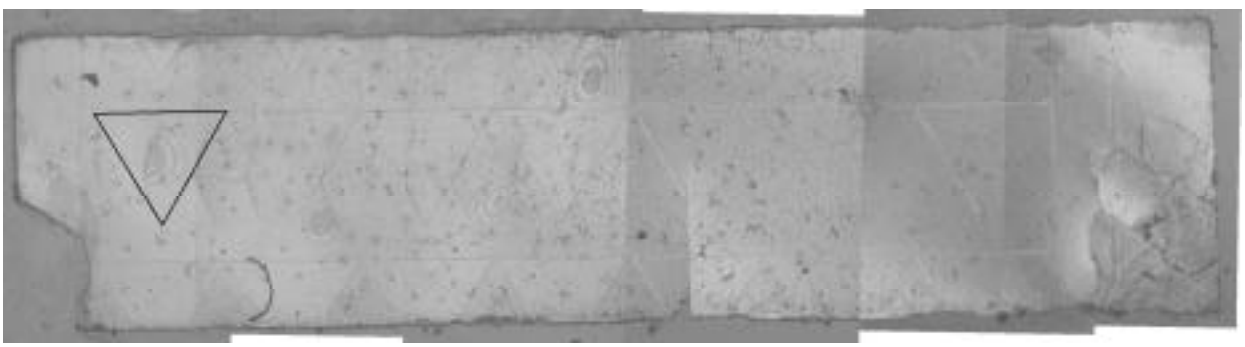

Figure 4: A top view of a CIS electro-optic scanner on z-cut $LiNbO_3$ The triangles are oppositely oriented domain states with a base of 1000 µm and height of 775 µm. The first triangle has been enhanced to show location. The sample curves up the right hand side.

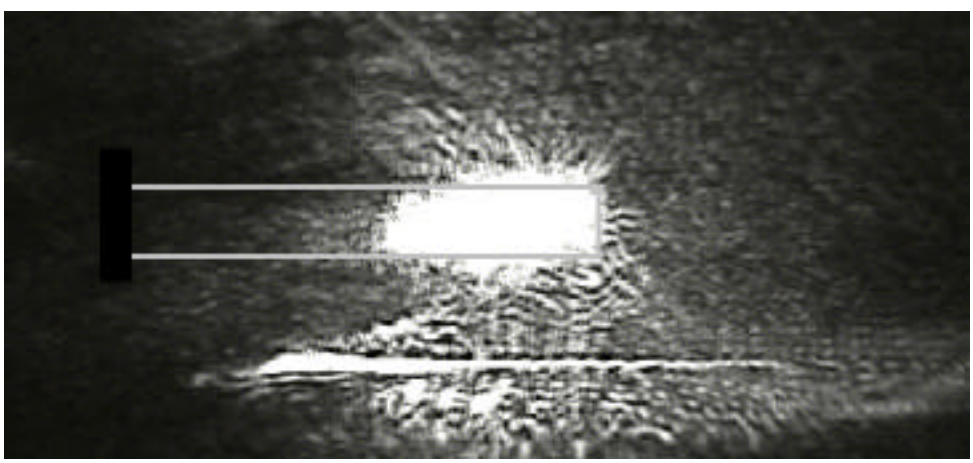

Figure 5: The output face of the crystal imaged in a CCD camera showing light coupling. The gray line shows position of device.

CONCLUSIONS

The successful lift off of a 2mm x 9 mm microengineered electro-optic device in $LiNbO_3$ was achieved by crystal ion slicing. The device slice measures ~6 μm thick. The predefined prismatic domain structures were preserved in the final device. Theoretically this device should operate at ± 90 V, which is significantly lower than the ± 4500 V required for a 0.3 mm thick bulk device. Optimal conditions for pre-liftoff annealing at 450°C for 40 s in flowing forming gas (5% H_2, 95% Ar) was found to have the shortest liftoff time and produced minimal etching of surface (~20 nm depth of etch lines). Light coupling was successfully achieved in device fragments. The scanner performace is yet to be characterized

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