# Optically reconfigurable ferroelectric metasurfaces

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**Abstract** We use ferroelectric-plasmonic metasurfaces to demonstrate volatile and non-volatile optical switching of near-infrared light. Plasmonic metasurfaces on lithium niobate enable high-contrast optical switching with ratios up to 2.37:1 (3.7 dB) due to photogalvanic and photorefractive effects, therefore rendering a compact platform for photonic computing. ©2022 The Author(s)

## Introduction

Photonic integrated circuits (PICs) and flat optics rely on passive optical elements made from high index dielectrics, (eg silicon and silicon nitride) or plasmonic metals.<sup>1,2</sup> Phase change materials,<sup>3</sup> nano-mechanical systems<sup>4</sup> and liquid crystals<sup>5</sup> have been the major material platforms to realise active nonlinearities and reconfigurable optical elements. However, such systems are difficult to incorporate with PICs. In this paper, we demonstrate a method for flat all-optical switches based on lithium niobate (LiNbO<sub>3</sub>) substrates. In we exploit photogalvanic and this work, photorefractive effects to develop all-optical switchable metasurfaces and functional films on LiNbO3 substrates.

## Photogalvanic and photorefractive effects

Bulk ferroelectric crystals have been used for a long time in nonlinear optics and electro-optics as materials with active tuning capabilities. Upon moderate laser power irradiation, these materials produce photo-excited carriers that can modify their optical properties. The charge carriers are separated by the net polarisation of the ferroelectric crystal and the thermal diffusion, therefore creating a space-charge distribution that modifies optical properties.<sup>6</sup> Here, we

present photonic metasurfaces and thin films adjacent to LiNbO3 substrates. The as-deposited films, due to charge screening, extract the charge carriers of undoped LiNbO3 (eg. oxygen and lithium vacancies) at the interface between gold and LiNbO<sub>3</sub>, while the post-annealing of the ITO/ LiNbO<sub>3</sub> samples introduces extra carriers, due to the thermal diffusion of tin inside the ferroelectric crystal. In the former case, carriers are mobile and induce reversible changes of the optical properties, while in the latter case deep carriers inside the bandgap of the crystal induce nonvolatile changes of the refractive index. These nanostructures serve as a flat platform with alloptical switching capabilities, necessary for the next generation of photonic architectures such as optical and neuromorphic computing.

## **Experimental setup**

The all-optical switching properties of the plasmonic-ferroelectric metasurfaces and films are evaluated using the pump-probe experimental configuration illustrated in Figure 1a. The pump laser is a diode laser at 520 nm and the probe laser is a diode laser at 1064nm. We perform lock-in detection of the probe beam at 5 kHz and record the laser signal with an InGaAs photodiode (PD). A pair of objectives are



Fig. 1: Measuring the all-optical switching of the ferroelectric metasurfaces and films. a) Schematic of the pump-probe experimental arrangement for transmission-mode measurements of metasurface sample nonlinear response. b) Photogalvanic charge separation under uniform illumination of a plasmonic metasurface. The arrows indicate the directions of the light-induced photogalvanic field and of the polar c-axis of the crystal. Annotations:  $\lambda/2$ : half-waveplate, Pol.: linear polariser, Obj.:objective, BS: beam splitter, HPF: high pass filter, PD: photodiode.



**Fig. 2 Au-LiNbO**<sub>3</sub> metasurface. (a) Oblique incidence and (b) cross-sectional scanning electron microscope images of a 700 nm period nano-hole array fabricated by focused ion beam milling in a 50 nm thick Au film on LiNbO<sub>3</sub>. Platinum covering the metasurface in b) is used for illustration reasons, scale bar: 350 nm. Inset: schematic of the cross-section annotating gold (Au) and lithium niobate (LiNbO<sub>3</sub>).

used to focus laser beams down to a spot size of 1  $\mu$ m. The k-vector of the probe beam is always vertical to the c-axis and the polarization vector lies on the same plane as the c-axis of the ferroelectric crystal. The polarization state of the probe laser is rotated with a linear polarizer (LP) and a half waveplate ( $\lambda$ /2). The probe power remains below 100  $\mu$ W, while the pump power is programmed to produce laser pulses of various power (order of few mW) and pulse widths.

## Samples fabrication

We use 100 µm thick, x-cut, congruent lithium niobate (LiNbO<sub>3</sub>) substrates. Then, we prepare two sets of samples. On the first set, we deposit on top of LiNbO3 a thin film of gold (Au) of subwavelength thickness, namely 50 nm, and we subsequently nanostructure an EOT (extraordinary optical transmission)-type metasurface7 with focused ion beam (FIB) milling, (acceleration voltage 30kV and ion beam current of 18pA). The design of the metasurface is a periodic array of nano-holes covering an area of 20 x 20 µm<sup>2</sup>. The period of the plasmonic metasurface is 700 nm and the diameter of each hole is 350 nm, see oblique view and crosssection scanning electron microscopy images on



Fig. 3 Optically reconfigurable ferroelectric metasurfaces and films. a) Reversible optical switching for two different probe polarization states enabled by the plasmonic metasurface. b) Two-level memory system of optical bits stored in the transmission level of the LiNbO<sub>3</sub> crystal. The green laser pulses of different duration and power perform the write and erase function, while the probe beam is used for the reading.

#### Figure 2.

The second set of samples is formed by depositing a thin layer of 50 nm of ITO on LiNbO<sub>3</sub> by electron-beam evaporation at room temperature. The as-deposited films are semi-transparent, therefore we subsequently anneal the samples at 500°C for one hour in ambient atmosphere to render the ITO transparent (T> 85% at 520 nm).

#### Results

The samples of the first set support reversible switching) transmission (volatile changes. Despite the polarisation insensitive metasurface design, the impact of the photogalvanic effect over the extraordinary refractive index of LiNbO3 is more sensitive than the ordinary refractive index to the green laser illumination. In Fig. 3a the optical power measured at the PD of the experimental setup of Fig. 1a is presented. This response is recorded for a pump power of 4 mW. Pump powers higher than 5 mW do not improve transmission contrast, as pyroelectric fields rise and reduce the modulation. However, no physical damage is recorded on the samples. The transmission contrast is defined as ILaser-on /ILaserand it reaches a maximum value of off

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the transmission levels can be understood as the generation of a space-charge along the pump beam. 8 Therefore, due to the linear electro-optic effect and the photo-elastic response of LiNbO<sub>3</sub>, the space-charge modifies the refractive index at the vicinity of the plasmonic metasurface. To evaluate the metasurface contribution of the switching contrast, we prepare a thin film which is transparent over the probe wavelength and lossy over the pump wavelength. For this, we have chosen to deposit a copper oxide film of 50 nm on the LiNbO3 substrates. We have also recorded a modulation of the probe beam, but a much higher pump power ~20mW induces a transmission contrast up to 1.25. Transmission contrast cannot be recorded for the bare LiNbO<sub>3</sub> substrate.

The second set of samples present non-volatile transmission changes. The transmission of ITO films on LiNbO3 upon two subsequent pulses of low and high power (of 5 mW and 20 mW, respectively) is presented in Fig. 3b. The first pulse (duration: 500 ms) can be considered as the writing pulse for the ON state, while the second pulse (duration: 5 s) is the erasing pulse that returns the transmission level back to the OFF state. The transmission contrast reaches 1.27 (defined by ILaser ON/ILaser OFF) as seen in Fig. 3b. There is a relaxation time of a few seconds until the transmission level stabilizes. This response is already considerable, as we have not developed yet a resonant metasurface on the ITO films. This behavior is similar to several photorefractive crystals and it is related to the motion of charges and the recombination of photo-carriers with unfilled deep traps. We anticipate that the fabrication of a photonic metasurface with an optical resonance at the probe wavelength will improve the switching contrast, while the switching dynamics could be engineered by an optical resonance at the pump wavelength.

# Conclusions

We have presented a novel method to perform all-optical switching in flat photonic architectures based on the combination of photonic metasurfaces and transparent conductive oxide films on LiNbO3 substrates. The reversible and irreversible transmission contrast between different types of samples paves the way for a new family of adaptive optical devices based on the LiNbO3 platform. The transmission contrast ratio of 2.37:1 demonstrated here is already adequate for short-reach (intra-/interchip) optical interconnect applications in data architectures.1 processing The functions

presented by plasmonic metasurfaces and functional films may form essential components such as optical modulators and optical storage elements towards the development of photonic computing. Furthermore, the rewritable optical properties could be used in planar switchable optical devices. Resonant structures could further improve the optical non-volatile switching in terms of speed, magnitude, and energy consumption.

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