

# Plasmonic Lithium Niobate Mach–Zehnder Modulators

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Cite This: *Nano Lett.* 2022, 22, 6471–6475



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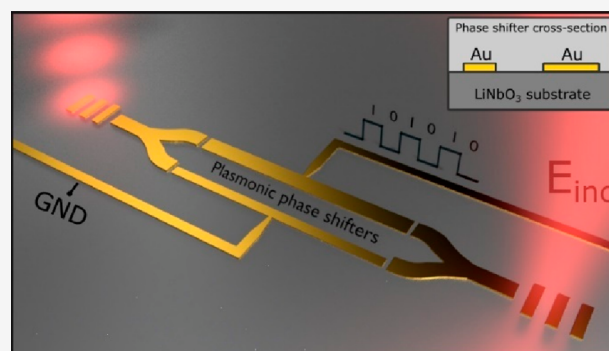
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**ABSTRACT:** Lithium niobate Mach–Zehnder modulators (MZMs) are present in a wide range of technologies and though fulfilling the performance and reliability requirements of present applications, they are becoming progressively too bulky, power inefficient, and slow in switching to keep pace with future technological demands. Here, we utilize plasmonics to demonstrate the most efficient ( $V_{\pi}L = 0.23$  Vcm) lithium niobate MZM to date, consisting of gold nanostripes on lithium niobate that guide both plasmonic modes and electrical signals that control their relative optical phase delay, thereby enabling efficient electro-optic modulation. For high linearity (modulation depth of  $>2$  dB), the proposed MZM inherently operates near its quadrature point by shifting the relative phase of the signal in the interferometric arms. The demonstrated lithium niobate MZM manifests the benefits of employing plasmonics for applications that demand compact ( $<1$  mm<sup>2</sup>) and fast ( $>10$  GHz) photonic components operating reliably at ambient temperatures.

**KEYWORDS:** Surface plasmon polaritons, Pockels effect, optoelectronics, nanophotonics, integrated optics



demand compact ( $<1$  mm<sup>2</sup>) and fast ( $>10$  GHz) photonic components operating reliably at ambient temperatures.

Mach–Zehnder Modulators (MZM) are key devices for electric-to-optical conversion in integrated photonics, telecommunications, and sensing.<sup>1–4</sup> In an integrated MZI modulator, a guided light beam is split into two paths and a phase difference between the two waveguide arms is induced using an electro-optically active material, most commonly lithium niobate (LN, LiNbO<sub>3</sub>) due to its preferential physical properties, such as moderate and stable electro-optic activity ( $\sim 30$  pm V<sup>-1</sup>) and wide optical transparency ( $0.35$ – $4.5$   $\mu$ m).<sup>5</sup> The phase difference in the waveguides results in a modulation of the output intensity by letting the phase-shifted beam interfere with a reference beam when the two waveguides are combined. Major performance limiting factors in terms of modulation bandwidth, modulation depth, energy per bit and footprint of an MZM are determined by the electro-optic efficiency of the phase-shifters.<sup>6</sup> As photonic technology utilizes poorly confined optical waveguides (titanium-indiffused or proton-exchanged in bulk LN)<sup>7–9</sup> which are limiting the electro-optic interaction, the efficiency of conventional MZM remains low. Thin-film lithium niobate modulators<sup>10–15</sup> lead to improvements in terms of compactness, operation speed and energy efficiency, while still demanding relatively long (on the mm scale) interaction lengths due to fundamental limitations in the achievable overlap between the optical and modulating electric fields.

Plasmonic lithium niobate technology<sup>16,17</sup> has recently emerged as a strong contender to overcome current performance limitations by providing electro-optic phase shifters with the highest efficiency ever reported in a lithium niobate device.

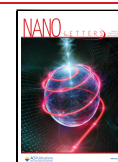
Because of the ultrafast Pockels effect (with a response time in the order of femtoseconds<sup>19</sup>) utilized in plasmonic lithium niobate devices with high modulation efficiency characterized by a voltage-length product as low as  $V_{\pi}L = 0.21$  Vcm, small device footprints with increased operation bandwidths far beyond 100 GHz are enabled. Among the demonstrated performance improvements in plasmonic lithium niobate devices, the implementation of an MZM which utilizes the extremely high modulation efficiency in plasmonic lithium niobate phase shifters remained unexplored and has yet to be demonstrated.

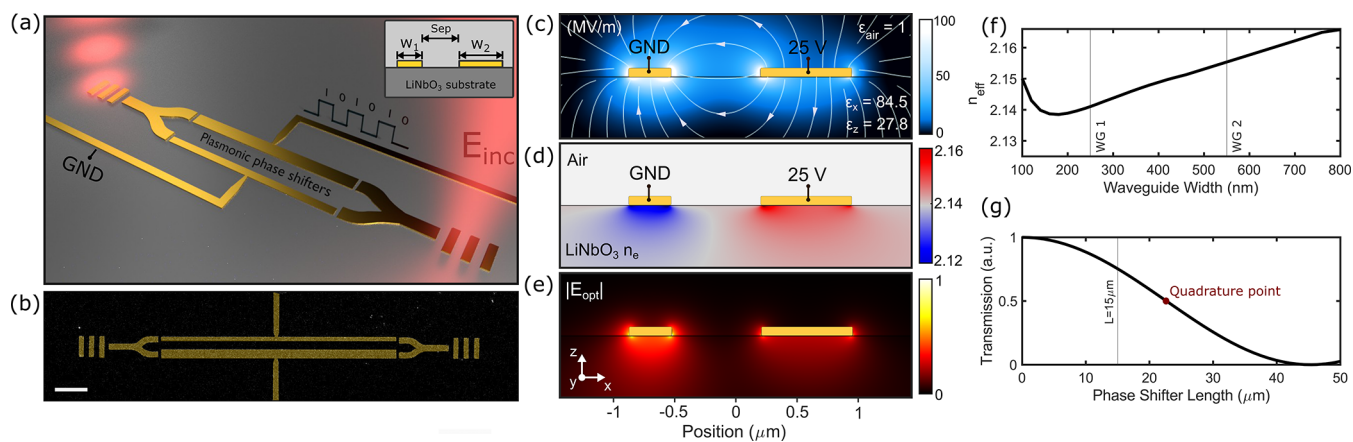
Here, we utilize the plasmonic lithium niobate platform to enable an efficient ( $V_{\pi}L = 0.23$  Vcm) and fast ( $\geq 10$  GHz) integrated MZM formed by phase shifters consisting of two only 15  $\mu$ m long plasmonic strip waveguides which supports both the optical signals converted to surface plasmon polaritons (SPPs) propagating at metal–dielectric interfaces and the electrical signal introducing the electro-optically induced phase shift.<sup>20–25</sup> The extreme confinement of the corresponding optical and electrostatic fields and the large overlap between the corresponding fields opens a path for novel highly efficient lithium niobate MZM.

**Received:** February 21, 2022

**Revised:** July 22, 2022

**Published:** August 11, 2022



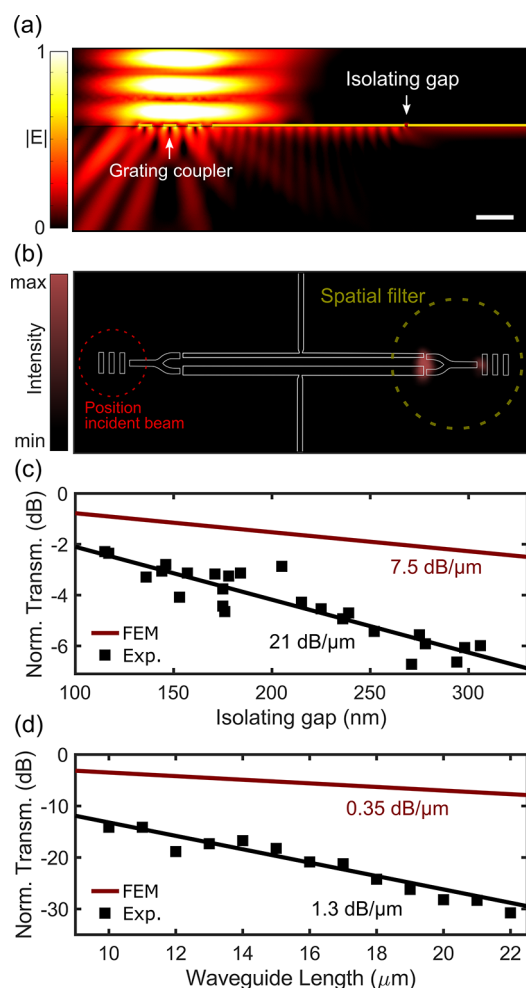


**Figure 1.** Plasmonic lithium niobate unbalanced MZM. (a) Conceptual image of electro-optic modulation in the MZM. The inset shows the cross-section of two parallel waveguides with the width of  $w_1 = 250$  nm and  $w_2 = 550$  nm, separated by 550 nm and placed on bulk z-cut lithium niobate (LN,  $\text{LiNbO}_3$ ) substrate. (b) False-colored scanning electron microscope (SEM) image of an investigated MZM. The scalebar represents 2  $\mu\text{m}$ . (c) Color-coded electric field and its contours upon applying a bias of 25 V. (d) Change of the extraordinary refractive index  $n_e$  due to the electro-optic (linear) Pockels effect in lithium niobate. (e) The optical field distribution of the antisymmetric mode overlaps with the electro-optically induced change of the refractive index in LN. (f) Mode effective index  $n_{\text{eff}}$  as a function of the waveguide width. (g) Transfer function for the MZM with 15  $\mu\text{m}$  phase-shifter length.

The studied asymmetric plasmonic lithium niobate MZM is based on two subwavelength gold nanowires as active phase shifter which are connected to two Y-branches serving as integrated waveguide splitter and combiner (Figure 1). Each gold nanowire supports the propagation of SPP modes guided along the nanowire with subwavelength field confinement,<sup>26–28</sup> thereby acting as an ultracompact waveguide in the nanophotonic integrated circuit. The plasmonic mode is launched by positioning a diffraction-limited beam ( $\lambda_0 = 1550$  nm) on the terminating metallic grating coupler with a calculated coupling efficiency of 10% at the wavelength of  $\lambda_0 = 1550$  nm. The mode is split by an integrated branch junction, which feeds the two active phase shifters by a 150 nm wide isolating air gap. The two 15  $\mu\text{m}$  long waveguides of the phase shifter section have different widths, that is, one waveguide has the width of  $w_1 = 250$  nm while the other has the width of  $w_2 = 550$  nm, and they are separated by edge-to-edge separation distance of  $g = 550$  nm. Because of the dependence of the waveguide geometry on the mode effective index of the supporting plasmonic mode (Figure 1f), the operation point of the MZM is passively shifted near the quadrature point (Figure 1g). This asymmetric configuration thereby offers high degree of modulation linearity at the operation regime of the MZM and the collapse of nonlinear distortions in the modulation spectrum. The refractive index of lithium niobate is modulated by applying a voltage across the two parallel wires comprising the plasmonic waveguides, and therefore the phase of the plasmonic modes propagating along them is shifted. Because of the Pockels effect in LN, the refractive index changes linearly to the applied electric field. The largest electro-optic coefficient  $r_{33}$  is along the optic axis of the LN crystal. Therefore, the electrical field  $E_z$  along the out-of-plane direction results in the strongest change of the extraordinary refractive index  $n_e$  in z-cut LN, according to  $\Delta n_e(E_z) \approx -0.5r_{33}n_e^3E_z$ . Considering the electric field distribution  $E(x, z)$  when the wires are biased (Figure 1c), the change of the refractive index is mainly located underneath the Au nanowires with an opposite modification between the individual wires due to an opposite direction of the electric field. The large overlap between the modification of the refractive index and the optical field distribution results in a

significant phase change with opposite directions in the two optical channels (push–pull phase modulation). Because of a different waveguide geometry, the strength of the electro-optic interaction is nonidentical for the two individual phase shifters. The optical mode and the electrostatic field are stronger confined in the narrower waveguide ( $w_1 = 250$  nm) resulting in a higher modulation efficiency, characterized by the calculated voltage-length product of 0.37 Vcm, compared to the wider phase shifter waveguide ( $w_2 = 550$  nm) exhibiting a voltage-length product of 0.57 Vcm. As the MZM inherently introduces phase shifts in push–pull operation, the voltage-length product of the device is reduced to 0.23 Vcm (Figure 1c–e). For the electrical isolation between the two individual phase shifter waveguides, isolating gaps between the phase shifters and the Y-branches are required. The separation between the waveguides is designed to be large enough to prevent parasitic directional coupling between the nonidentical waveguides (Supporting Information Note S2). Furthermore, the isolating gap between the phase shifters and the Y-branch is small enough ( $g \ll \lambda_{\text{SPP}}$ ) to allow optical transmission through the air gap. The scattering losses are characterized as a function of the gap size (Figure 2). For the nominal gap size of 150 nm, which is much smaller than the wavelengths  $\lambda_{\text{SPP},1} = 725$  nm ( $w_1 = 250$  nm) and  $\lambda_{\text{SPP},2} = 720$  nm ( $w_2 = 550$  nm) of the SPPs propagating along the phase shifters, an optical loss of  $\sim 3.5$  dB per isolating gap is estimated. Furthermore, the propagation loss is investigated by an array of single waveguides with different length. The propagation loss in a 15  $\mu\text{m}$  long and 350 nm wide waveguide is estimated to be 19.5 dB, which indicate much higher losses than expected from simulations (0.35 dB/ $\mu\text{m}$ ) and are likely attributed to additional losses due to the titanium adhesion layer and the grain boundaries of the polycrystalline gold nanowires.

The proposed MZM device is characterized by positioning a focused linearly polarized beam ( $\lambda_0 = 1550$  nm) onto the grating coupler at normal incidence (Supporting Information Note S1). The SPPs are launched and feed the phase shifters at the interferometric arms of the MZM before the signals interfere at the opposite Y-branch. For obtaining the Mach–Zehnder transfer function, the scattering signal of the output



**Figure 2.** Investigation of the optical losses in a single plasmonic waveguide consisting of a 50 nm thick stripe and an air gap for electric isolation. (a) Electric field distribution simulated for a waveguide with grating coupler and isolating gap of 100 nm. (b) Experimental optical far-field image captured by an infrared camera with a spatial filter at the output port. The laser beam ( $\lambda_0 = 1.55 \mu\text{m}$ ) is positioned on the left grating coupler with its reflection being blocked in front of the camera to prevent image overexposure. The structure design is superimposed as a guide to the eye. (c) Measured normalized transmission as a function of the width of the isolating gap. (d) Measured waveguide transmission as a function of the waveguide length without an isolating gap.

grating is spatially filtered and detected with an infrared photodiode. The voltage-dependent output intensity follows the expected transfer function of an unbalanced MZM exhibiting a passive phase shift of  $0.28\pi$  (Figure 3a). Continuous, robust, and quasi-linear intensity modulation between  $-25 \text{ V}$  and  $+25 \text{ V}$  is observed with a maximum modulation depth of 2.5 dB. The significant modulation in the ultracompact MZM characterized by the voltage-length product of 0.23 Vcm represents to-date the highest modulation efficiency in a lithium niobate MZM (Supporting Information Note S3). Dielectric breakdown along the isolating nanogaps restricts the maximum applicable voltage which limits the achievable depth of modulation but can potentially be fully avoided by interfacing the plasmonic phase shifters with dielectric nanophotonic waveguides.<sup>23,24,29</sup> Capitalizing on the wide optical transparency of lithium niobate and the relatively low plasmonic losses at infrared wavelengths, spectrally

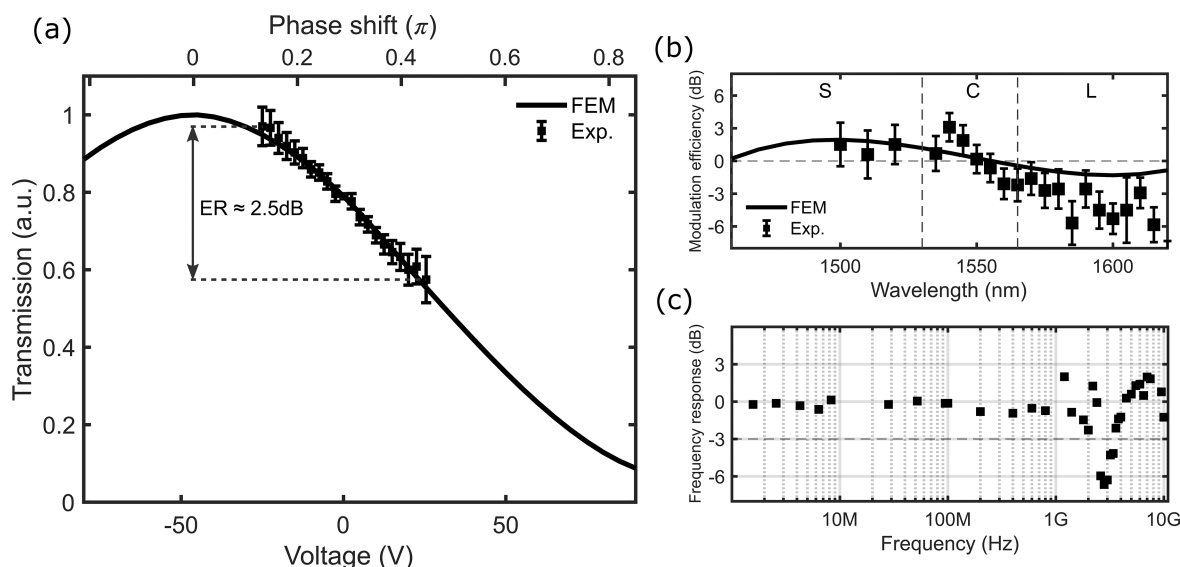
broadband operation over the telecommunication wavelength bands S, C, and L (1500–1600 nm) with  $<6 \text{ dB}$  modulation depth variation is demonstrated. The observed variations are caused by the wavelength dependence of the electro-optic field overlap and the shift of the operation point in the transfer function.<sup>17</sup> The electro-optic frequency response of the MZM is characterized from 1 MHz to 10 GHz (Figure 3c). The device exhibits a flat frequency response except a dip of  $-7 \text{ dB}$  at the frequency  $2.8 \pm 1 \text{ GHz}$  which is likely to be induced by nonideal impedance-matching of the used electrical feedline. The calculated device capacitance of only 3.3 fF indicates potential operation at frequencies above 900 GHz at 50  $\Omega$  resistive load ( $f = 1/[2\pi RC]$ ), which meets the future bandwidth demands for optical integrated circuits.

## DISCUSSION

By utilizing plasmonic lithium niobate technology, we demonstrated a broadband Mach–Zehnder modulator featuring a half-wave voltage-length product of 0.23 Vcm, which to date is the lowest value for a lithium niobate MZM. Spectrally broadband operation with  $<6 \text{ dB}$  modulation depth variation over the optical telecommunication wavelengths (1500–1600 nm) and high-speed operation up to 10 GHz (with a potential bandwidth of 900 GHz) was demonstrated. Highly linearity modulation response is realized by passive control of the relative phase in the nonidentical waveguides forming the interferometric arms. Optical transmission and scattering losses introduced by the isolating air gap have been characterized and can potentially be further reduced by using a high-quality (monocrystalline) Au platform for the plasmonic phase shifters<sup>18</sup> and by eventually interfacing the plasmonic waveguides with a low-loss photonic circuit.<sup>24</sup> As lithium niobate provides the critical material properties for practical deployment in electro-optic modulation applications, it remains the material-of-choice for telecommunication technology. By demonstrating the possibility of realizing a lithium niobate MZM that harvests the attractive feature of plasmonics to guide both coupled electromagnetic modes and electrical signals that control their phase shift in the same metal circuitry, we enabled an exceptionally compact and efficient MZM with potential applications in next generation high speed electro-optics that can fulfill the performance-demanding needs of future data processing and quantum technologies.

## METHODS

**Simulations.** Numerical simulations of the electro-optic response, the optical losses and the coupling efficiency of the MZM were carried out using a commercial finite element method solver (Comsol Multiphysics 5.5). For the electrostatic simulations, the cross-section of the two phase shifters is modeled with the relative permittivity  $\epsilon_{\text{air}} = 1$  of air and the strain-free static relative permittivity tensor of lithium niobate ( $\epsilon_{xx} = \epsilon_{yy} = 27.8$ ,  $\epsilon_{zz} = 84.5$ ) taken from Jazbinsek et al.<sup>30</sup> The electrostatic field distribution  $E(x,z)$  in the phase shifter cross-section is calculated with two gold electrodes and set to ground and  $V_{\text{bias}}$ . The electro-optically induced birefringence in LN is calculated by using the Pockels coefficients from Jazbinsek et al.<sup>30</sup> (with the diagonal terms, i.e.,  $\Delta n_{ii} = -0.5r_{iiz}n_{ii}^3E_z$  with  $r_{xxx} = r_{yyy} = 10.12 \text{ pm/V}$  and  $r_{zzz} = 31.45 \text{ pm/V}$ ). The optical mode analysis is conducted by considering the lateral distribution of the electro-optically modified refractive index of LN, whereas the unmodified refractive indices of Au and LN are taken from



**Figure 3.** (a) Measured transfer function of the unbalanced Mach–Zehnder modulator, normalized by the measured transmission at 0 V. Error bars represent standard deviations and are deducted from repeated measurements. For the measured voltage range of  $\pm 25$  V, an extinction ratio (ER) of 2.5 dB was measured. (b) Wavelength dependence of the modulation efficiency (at a driving voltage of  $V_{AC} = 15$  V) normalized by the modulation efficiency measured at the free-space wavelength of 1550 nm. Error bars represent standard deviations. (c) Measured frequency response as a function of the RF modulation frequency.

Johnson and Christy<sup>31</sup> and Zelmon et al.<sup>32</sup> ( $n_{xx} = n_{yy} = n_o = 2.211$ ,  $n_{zz} = n_e = 2.138$  at  $\lambda_0 = 1550$  nm). Possible losses introduced by imperfect Y-splitting elements are neglected and a splitting ratio of 0.5/0.5 is assumed.

**Sample Fabrication.** The Mach–Zehnder modulators are fabricated on commercially available z-cut lithium niobate substrates by electron beam lithography (using a scanning electron microscope JEOL JSM-6490LV with an acceleration voltage of 30 keV) in spin-coated 250 nm thick PMMA positive resist and a 40 nm thick conductive polymer layer (AR-PC 5090, Allresist) which serves as a metallic charge dissipation layer during the writing (electron doses varying between 200 and 250  $\mu\text{C}/\text{cm}^2$ ). After resist development, the devices are formed by depositing a 3 nm titanium adhesion layer and a 50 nm gold layer by thermal evaporation and subsequent 8 h lift-off. RF feed lines are patterned beforehand on the LN chip by shadow masks metal deposition (5 nm Ti/100 nm Au).

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.2c00714>.

Optical setup for the electro-optic characterization of the plasmonic Mach–Zehnder modulators, coupled-mode theory analysis for the investigated two-wire transmission line, comparison with state-of-the-art electro-optic Mach–Zehnder modulators (PDF)

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## Notes

The authors declare no competing financial interest.

## ■ ACKNOWLEDGMENTS

The authors gratefully acknowledge the support from the Villum Kann Rasmussen Foundation (Award in Technical and Natural Sciences 2019).

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