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Harnessing nonlinearities near material absorption resonances for reducing losses in plasmonic modulators

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Abstract: The electro-optic coefficient (Pockels coefficient) is largest around the absorption resonance of a material. Here, we show that the overall losses, the power consumption and the footprint of plasmonic electro-optic modulators can be reduced when a device is operated in the vicinity of absorption resonances of an electro-optical material. This near-resonant operation in plasmonics is contrary to what is known from photonics where off-resonant operation is required to minimize the overall losses. The findings are supported by experiments demonstrating a reduction in voltage-length product by a factor of 3 and a reduction in loss by a factor 2 when operating a plasmonic modulator near resonance compared to off-resonant.

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1. Introduction

Electro-optic (EO) modulators are key for realizing highly integrated electro-optic circuits. In such circuits, fast and efficient light modulation based on linear and nonlinear electro-optic effects are essential to realize high-speed operation with a high-integration density [1, 2]. Further, downsampling of optical communication systems for on-chip applications poses strict restrictions for the maximal tolerable energy dissipation to keep heat dissipation below critical levels [3]. However, to achieve this ambitious goal, EO modulators need to become more efficient in terms of the required driving voltage [4], while retaining high operating speed across a large optical spectral range.

To accommodate such stringent requirements in speed, compactness, energy efficiency and driving voltage, current research focuses on a variety of approaches. These include novel electro-optics modulation mechanisms based on 2D materials [5], quantum effects [6–8], or the introduction of structural resonances that enable an enhancement of the underlying electro-optic effect [9]. The latter approach uses disc or ring resonators to build an optical cavity in order to enhance the free-carrier dispersion (FCD) effect in silicon [9–11]. Non-
resonant modulators based on the FCD require large driving voltages to achieve sufficient modulation across mm-long interaction lengths [1,12,13]. By introducing resonant approaches with high Q-cavities it is possible to enhance the FCD-effect so that cavity lengths of 10s of microns are sufficient to reach sub-V driving voltages [10,14]. However, large Q-factors are generally narrowband (tens of GHz) and energy intensive thermal tuning of the resonators is required [11]. An alternative that circumvents the speed limitation is given by plasmonics [15,16]. In plasmonic approaches, light is guided at metallic-dielectric interfaces and it is the metal that provides a material built-in resonance which enables the enhancement of nonlinear effects by confining the light to sub-diffraction limited areas [17]. The resonance is caused by the free electron cloud in metals, which couples with the electromagnetic wave to form so-called surface-plasmon polaritons (SPPs) [18,19]. SPPs have been utilized successfully in FCD plasmonic modulators [20–23] and plasmonic organic hybrid modulators [24–27]. The latter technology uses the Pockels effect in organic electro-optic (OEO) materials [26,28–30]. To date, the plasmonic enhancement has led to a more than 10-fold reduction of the voltage-length ($U/L$) product compared to the best photonic approaches [31–33].

While the voltage-length product has been enhanced by the resonance of the passive metal, the active OEO materials still have an untapped potential. For instance, the Pockels effect increases when operating the devices close to the OEO-material absorption lines. Thus, the performance of such modulators is ultimately defined by a trade-off between nonlinearities and optical absorption [34]. Compared to photonic modulators, which are typically 100s of microns long, plasmonic modulators are only several microns long and increased material absorption might not be relevant for the overall performance.

In this study, for the first time, we investigate the possibility to enhance the performance of plasmonic-organic hybrid (POH) modulators by operating the devices in the vicinity of the OEO material’s resonance. We theoretically and experimentally demonstrate that the performance of POH modulators can be enhanced by up to 100% without increasing optical losses when adapting the operation wavelength to exploit the material resonances. In terms of voltage-length product an even greater performance enhancement of 200% is demonstrated. This approach opens a way for reducing the energy consumption of future POH modulators by five times, without increasing optical losses, while maintaining a compact footprint and an EO bandwidth beyond 170 GHz.

2. Plasmonic-organic hybrid modulators

The concept of plasmonic-organic hybrid (POH) modulators [24,25] can be best understood with the help of Fig. 1(a) showing a top view of a plasmonic phase modulator (PPM).

![Fig. 1. a) Colorized SEM images showing the top view of a plasmonic organic hybrid (POH) phase shifter of length $L$. The phase shifter consists of a plasmonic metal-insulator-metal slot waveguide. Si waveguides are used to feed and extract light to and from the phase shifter. (b) An artist’s cross-section view of the phase shifter. The organic electro-optic material (HD-BB-OH/YLD124) fills the slot. A voltage applied to the electrodes changes the phase of SPPs propagating in the slot due to the Pockels effect. Image adapted from [35].](image-url)
The modulator comprising of silicon (Si) access waveguides (blue) and a plasmonic slot waveguide formed by two gold (Au) contact pads (yellow). The slots of width \( w \) are typically 40 nm- to 160 nm-wide and are filled with a binary chromophore composite of 75% HD-BB-OH and 25% YLD124 [36], see Fig. 1(b). This composite features in-device EO coefficients of up to 325 pm/V and is the outcome of theory guided OEO material design [36]. This progress has been accompanied with an increase of the temperature stability of binary and monolithic OEO materials [35,36] tending towards the impressive \( T_g \) values of guest-host materials such as SEO100 and M3, see Table 1. The new OEO material has not yet been fully investigated for thermal stability. Still, some simple thermal stability tests have been performed, showing that the electro-optic coefficient drops by 15% after a 3h exposure to 80°C in vacuum. Exposure of additional 3h at 80°C in ambient atmosphere does not cause a further decrease \( r_{33} \).

| Table 1. – In POH modulators demonstrated performance of organic electro-optic materials |
|---------------------------------|---|---|---|---|---|
| OEO Material                  | Year | \( r_{33} \) thin-film (1310 nm) [pm/V] | \( r_{33} \) in-device (1310 nm) [pm/V] | \( r_{33} \) in-device (1550 nm) [pm/V] | \( T_g \) [°C] | Ref |
| GigOptix M3                   | 2014 | ~90 | - | 21 | ~170 | [25,37] |
| Soluxra SEO100                | 2015 | 110/260\(^a\) | - | 70 | ~150 | [38] [25] |
| 75%PSLD41/25%YLD124           | 2015 | 285 | - | 116 | ~97 | [25,39] |
| DLD164                        | 2015 | 137 | - | 180 | ~66 | [40], [24] |
| JRD1                          | 2016 | 343/556\(^a\) | - | 193 | ~82 | [41], [35] |
| 75%HD-BB-OH/25%YLD124        | 2017 | 253 | 290 | 220 | ~110 | [36] [this work] |

\(^a\) larger EO coefficient demonstrated in SOH devices, see Table 1 in [35]. \(^b\) with charge blocking layer. \(^c\) measured at 1600 nm, larger \( r_{33} \) is expected at lower wavelengths. Table adapted from [35].

The operation principle depicted in Fig. 1(a) is as follows. TE-polarized light from the input Si waveguide is converted by a tapered structure to surface-plasmon-polaritons (SPPs) propagating in the Au slot waveguide [42]. The Pockels effect in the OEO material is used to encode an electrical signal on the phase of the propagating SPPs. Finally, the phase-modulated SPPs are converted back to photonic modes of the output silicon waveguide by the second taper structure. SPPs - such as those propagating in the plasmonic slot waveguide - have the unique property to confine electromagnetic energy below the diffraction limit. However, the tight confinement comes at the price of increased propagation loss. In order to limit the effect of plasmonic losses, phase shifters need to be efficient so that the accumulated losses are low. Due to their high efficiency, plasmonic phase shifters are short and come with typical lengths between 5 \( \mu \)m [24] to 50 \( \mu \)m [43] depending on the application. Short phase shifters (<5 \( \mu \)m) display low optical losses, but require relatively high \( U_x \) voltages of 10 V causing an electrical energy consumption of 25 fJ/bit [31]. Contrarily, long phase shifters (>10 \( \mu \)m) enable reduced \( U_x \) voltages at the cost of higher modulator losses [44–46]. A way to reduce the driving voltage without suffering from higher losses is to make use of material resonances given by the OEO-material and by gold. These resonances cause a wavelength dependency of the linear and nonlinear optical properties of the materials, which allows further optimization of the POH technology as will be discussed in the following section.

3. Enhancing the efficiency by material resonances

The efficiency (\( \eta \)) of photonic and plasmonic (e.g. POH) phase modulators is generally expressed by the ratio between phase shift (\( \Delta \beta = k_0 \Delta n_{\text{eff}} \)) and attenuation coefficient (\( \alpha_{\text{coeff.}} = 2k_0 \Delta n_{\text{eff}} \)) [15]. The phase shift is induced by the Pockels effect and is expressed by the free-space wave vector (\( k_0 = 2\pi/\lambda \)) times the change of the effective refractive index (\( \Delta n_{\text{eff}} \)). Losses are given by the free-space wave vector times the imaginary part of the effective
refractive index ($n_{\text{eff}}$). For a first qualitative discussion we can approximate (see Appendix) the efficiency by the following expression.

$$\eta(\lambda) = \frac{\Delta n_{\text{eff}}(\lambda)}{n_{\text{eff}}(\lambda)} = \frac{n_{\text{OEO}}(\lambda)^3 \cdot r_{33}(\lambda)}{(n_{\text{OEO}}(\lambda) + n_{\text{Metal}}(\lambda))},$$

(1)

where $\lambda$ is the wavelength. The nonlinear coefficient is $r_{33}$. The material properties are given by the real part ($n_{\text{OEO}}$) and the imaginary part ($n'_{\text{OEO}}$ and $n''_{\text{Metal}}$) of the refractive index. The denominator highlights the loss trade-off between metals and OEO materials. Losses are dominated by the metal when operating far off the OEO material’s resonance ($\lambda >> \lambda_{\text{resonance-OEO}}$), while losses are dominated by the OEO in close vicinity to the resonance. In order to enhance the efficiency, the ratio in Eq. (1) needs to be maximized. Both the refractive indices and the electro-optic coefficient are subject to dispersion which provides a lever to enhance the efficiency of plasmonic phase shifters [34].

Material dispersion affects both the real ($\Delta n_{\text{eff}}$) and imaginary ($n''_{\text{eff}}$) part of the effective refractive index, and it is strongest in the vicinity of the absorption peaks, as shown in Fig. 2(a) for the case of the OEO-material HD-BB-OH/YLD 124. The exact numerical values are stated in Table 2. The plot shows ellipsometry measurements of the OEO-material over the wavelength range from 0.5 $\mu$m to 1.75 $\mu$m for both the real part (blue) and imaginary part (red).

![Graph showing optical properties of the OEO-material as a function of wavelength.](image)

- Fig. 2. Optical properties of the OEO-material as a function of wavelength. (a) The measured real (blue) and imaginary (red) parts of the effective refractive index are plotted in the wavelength range from 0.5 $\mu$m to 1.65 $\mu$m, see Data File 1 for underlying values. The bluish area to the right of the absorption highlights the area where losses are low and the real part of the electro optic coefficient is high. This area is most promising for optimizing the efficiency of plasmonic modulators. (b) The electro-optic coefficient ($r_{33}$), which is a measure of the strength of the material’s nonlinearity, is plotted versus wavelength. The nonlinearity enhances by up to a factor 3 upon approaching the absorption resonances. (c) The absorption coefficients versus wavelength for plasmonic (orange) and photonic (green) waveguides compared to the contribution from the OEO-material losses. It can be seen that the photonic waveguide losses are dominated by the losses of the OEO-material. In long photonic waveguides it thus is necessary to operate devices at long wavelengths. In contrast material losses hardly contribute to the overall losses of plasmonic waveguides and it thus is favorable to operate them at shorter wavelengths.

Absorption starts to emerge at the OEO material’s bandgap at 1075 nm (vertical dashed line) as photons can be absorbed to excite electrons from the ground state $|g\rangle$ to the excited state $|e\rangle$ of the OEO-molecule. This not only causes a change in the linear optical properties ($n'_{\text{OEO}}, n''_{\text{OEO}}$) but also results in a resonant enhancement of the Pockel’s effect [47], see Fig. 2(b). Its strength is represented and quantified by the tensor of the electro-optic coefficient ($r$), whereas in the case of OEO-materials the $r_{33}$ component is dominant [48]. The resonant
enhancement of the electro-optic coefficient in the vicinity of the absorption can be estimated with the help of a two-state model as introduced in the work [47]. Figure 2(b) shows the electro-optic coefficient $r_{33}$ over a wavelength range from 1.7 μm down to 1.2 μm. The electro-optic coefficient enhances by more than three times when approaching the material resonances. Contrarily, $n''_{\text{OEO}}$ increases by almost four orders of magnitude in the vicinity of the absorption lines over the same wavelength range, see Fig. 2(c). Thus, it is important to note that the optimal efficiency of phase modulators is not necessarily achieved at the wavelength of maximal nonlinear response, but rather off-resonance, as shown in [34]. In that study, Mossman et al. assume that losses are mainly caused by the OEO material; however, in real devices, losses are dominated by waveguide roughness [49] and/or plasmonic losses [24].

These losses are shown in Fig. 2(c) with the help of the effective refractive index’s imaginary part $(n')$ as a function of the wavelength for photonic (green) and plasmonic (orange) waveguides and should be compared to the imaginary part of the OEO material’s refractive index $(n_{\text{OEO}})$ serving as a reference. The solid green line shows the estimated propagation losses of silicon-organic hybrid (SOH) modulators [50] assuming that the photonic slot waveguides are filled with HD-BB-OH/YLD 124. In these structures, losses are dominated by the OEO material even far off-resonance. The dashed green line serves as a reference for the measured propagation losses at 1550 nm, which arise solely from sidewall roughness [49].

Losses of POH modulators are represented by the orange lines. The dashed line displays the calculated plasmonic loss of an 80 nm wide slot waveguide filled with an absorption-free OEO material. Thus, absorption only originates from ohmic/plasmonic losses and only a slight increase of 15% can be observed when reducing the wavelength from 1.6 μm to 1.2 μm. In contrast, as discussed above, the absorption of the OEO material increases by multiple orders of magnitude when reducing the wavelength. Calculating the plasmonic losses while taking into account the losses of the OEO material shows (solid orange line) that the OEO material is only dominating losses for wavelengths below 1.15 μm $(n''_{\text{OEO}} > n''_{\text{Metal}})$.

In summary, we have shown that photonic modulators utilizing HD-BB-OH/YLD 124 would be best operated far off the material’s resonance frequency ($\lambda > 1.5$ μm). Instead, POH modulators can be operated in closer vicinity of the OEO material ($\lambda > 1.2$ μm) without increasing the modulator loss. This is a strong advancement towards efficiency enhancement, as the electro-optic (nonlinear) response increases when getting closer to the absorption wavelength of the material. As a consequence, the nonlinear response enhances by 60% when operating at 1.25 μm instead of 1.6 μm, while losses do not increase significantly.

4. Simulation of the POH modulator performance enhancement

In the previous section, we have discussed the potential of OEO materials to enhance the efficiency of plasmonic organic hybrid modulators. In this section, we will discuss how the design of the plasmonic waveguide can be tailored to further increase the efficiency. For this purpose, we provide a separate analysis of how $\Delta n'_{\text{eff}}$ and $n''_{\text{eff}}$ depend on the geometry, the plasmonic dispersion relation and the material dispersion of the OEO material.

The effective refractive index change is subject to the dispersion of the OEO material and the geometry of the waveguide [24]

$$\Delta n_{\text{eff}} = \Gamma(\lambda) \cdot n_{\text{slow}}(\lambda) \cdot \Delta n_{\text{OEO,rel}}(\lambda). \quad (2)$$

Here, $\Gamma$ is the field-energy interaction factor defined by the geometry, $n_{\text{slow}}$ is the slowdown factor which is due to the plasmonic dispersion relation, and the relative change of the material’s refractive index is $\Delta n_{\text{OEO,rel}}(\lambda)$, which is due to the material dispersion of the OEO-material. This refractive index change is given by $\Delta n_{\text{OEO,rel}}(\lambda) = n''_{\text{OEO}}(\lambda) \cdot r_{33}(\lambda) / 2 \cdot U / w_{\text{slot}}$, where $r_{33}$ is the electro-optic coefficient. The electric field, which causes the phase shift, is defined by an applied signal voltage $(U)$ dropping across the slot width $(w_{\text{slot}})$. The voltage $U$
is fixed to 1 V throughout the simulations, however, any voltage could be assumed for simulation. During device operation and poling higher voltages are applied to the device.

Figure 3 shows the simulated wavelength dependence of the field-energy interaction factor $\Gamma$, Fig. 3(a), of the slow down factor $n_{\text{slow}}$, Fig. 3(b), and of the relative refractive index change $\Delta n_{\text{OEO,rel.}}$ in Fig. 3(c), for slot widths of 50 nm (blue), 100 nm (green), 150 nm (yellow) and 200nm (red) for a plasmonic slot waveguide, see Fig. 1. A slot height of 150 nm and a sidewall angle of 5 degrees are assumed throughout simulations. The permittivity of Au is taken from [24], the refractive index of the OEO material HD-BB-OH was obtained from ellipsometry measurements, see Data File 1. The electro-optic coefficient is 180 pm/V at 1.6 $\mu$m similar to DLD-164. The refractive index of SiO$_2$ is based on ref [51].

![Figure 3. Wavelength-dependent contributors to the modulator efficiency for various slot widths.](image)

(a) Field energy interaction factor $\Gamma$, (b) slow-down factor $n_{\text{slow}}$ due to plasmonic dispersions and (c) relative change in the OEO-refractive index due to eoe material dispersion.

In the following we will discuss the individual components and give a brief interpretation of their physical meaning. A mathematical derivation and more detailed explanation of the significance of $\Gamma$ and $n_{\text{slow}}$ can be found in [24]. In Fig. 3(a), the field-energy interaction factor $\Gamma$ shows almost no wavelength dependency. This quantity describes how well the electrical rf-field and the electrical optical field is confined to the electro-optical material in the slot. Better confinement results in a larger nonlinear interaction. The weak wavelength dependence can be attributed to two effects compensating each other: on the one hand, shorter wavelengths enable a tighter vertical confinement of the plasmonic mode; on the other hand, when reducing wavelength, gold becomes less metallic and more field is leaking horizontally into the metal. Figure 3(b) shows the slow-down factor. This factor is larger for shorter wavelengths and narrower slots. A high value for this factor is beneficial for the efficiency of the modulator, as slowed-down light experiences a longer interaction time with the OEO material. The slow-down effect is due to stronger coupling of light to the free charge-carriers in the metal when the slot is narrowed down or when the wavelength is reduced [19]. Finally, Fig. 3(c) highlights the influence of the OEO material. The material’s dispersion causes both the refractive index as well as the electro-optic coefficient to increase with shorter wavelength, resulting in an increase of the material’s relative refractive index change $\Delta n_{\text{OEO,rel.}}$ up to a factor 3.
Fig. 4. The normalized change of the effective refractive index’s real part (a), the effective refractive index’s imaginary part (b), and the change in the phase shifter’s efficiency $\eta$ (c) are shown. The phase shifter efficiency $\eta$ is the ratio of $\Delta n_{\text{eff}}$ and $k_{\text{eff}}$. By getting closer to the resonance frequency the efficiency enhances up to two times before losses start to dominate.

The combination of those three effects results in a wavelength-dependent change of the effective refractive index, as shown in Fig. 4(a). The effective refractive index change is normalized to a wavelength of 1.55 $\mu$m for all depicted slot widths. The plot shows a 3-fold enhancement of $\Delta n'_{\text{eff}}$ when reducing the operation wavelength of the modulator from 1.55 $\mu$m to 1.2 $\mu$m. Another important aspect are the optical losses $n_{\text{eff}}$ which are shown in Fig. 4(b). Their values have been normalized to the value at 1.55 $\mu$m. We can observe that losses only increase slightly over the broad spectrum ranging from 1.65 $\mu$m to 1.25 $\mu$m. At approximately 1.25 $\mu$m, absorption within the OEO-material starts to dominate and a steep increase of $n'_{\text{eff}}$ can be observed. For narrower slots the absorption of the OEO material starts dominating at shorter wavelengths, as plasmonic losses are larger in narrow slots [35]. The efficiency $\eta$, which is defined as the ratio of the two competing parameters $\Delta n_{\text{eff}}$ and $n_{\text{eff}}$, is shown in Fig. 4 (c). The different wavelength dependencies of both result in a maximum of $\eta$ around 1.25 $\mu$m, indicating a potential optimum operation wavelength of POH modulators using HD-BB-OH/YLD-124. At this wavelength the modulators can be operated with a 75% to 110% higher efficiency.

In the previous paragraph the optimal operation point was found based on the modal properties of SPPs in plasmonic slot waveguides ($n_{\text{eff}}$ and $\Delta n_{\text{eff}}$). However, in practical devices performance is reported in terms of the voltage-length product ($U_{\pi}L$). The former is defined as the voltage required to induce a phase shift of $\pi$ when applied to a phase modulator of length $L$. It is related to the modal properties by the relation [35]:

$$U_{\pi}L = \frac{\lambda \cdot W_{\text{slot}}}{\Gamma(\lambda) \cdot n_{\text{slow}}(\lambda) \cdot n_{\text{OEO}}(\lambda) \cdot r_{\text{sl}}(\lambda)}.$$  \hspace{1cm} (3)

However, the voltage-length product only describes the strength of the light-matter interaction. Instead, the loss-voltage-length product ($\alpha U_{\pi}L$) can be applied to obtain a figure of merit (FOM) equivalent to the efficiency $\eta$. This FOM is defined by the strength of the light-matter interaction ($U_{\pi}L$) times the associated optical propagation loss ($\alpha$) in dB per micron. In this definition the propagation loss per micron ($L = 1$ $\mu$m) are related to $n'_{\text{eff}}$ by

$$\alpha = 10 \cdot \log_{10} \left( \exp \left( \frac{4\pi}{\lambda} n'_{\text{eff}} \cdot L \right) \right).$$  \hspace{1cm} (4)

Figure 5(a), (b) and (c) respectively show the absolute values of $U_{\pi}L$, the propagation loss and the values of $\alpha U_{\pi}L$ as a function of wavelength, for four different slot widths.
Fig. 5. The voltage-length product ($U_\pi L$) (a), the propagation loss (b) and the loss-voltage-length product ($\alpha U_\pi L$) (c) as a function of wavelength for various slot widths of 200 nm (red), 150 nm (orange), 100 nm (green) and 50 nm (blue). The decrease in the voltage length product (a) overcompensates losses as long as the propagation losses (b) are not dominated by the absorption in the OEO-material ($\lambda >1.2 \mu m$). This results in a maximal efficiency of the modulators in terms of $\alpha U_\pi L$ between 1.2 $\mu m$ and 1.25 $\mu m$. The optimal point of narrower slots are blue shifted resulting in a larger light-matter interaction. Further, the enhanced light matter interaction for narrower slots (25 $V_{\mu m}$ for 50 nm compared to 114 $V_{\mu m}$ for 200 nm) are overcompensating the increased losses caused by narrower slots. This results in $\alpha U_\pi L$ of 25 dBV for a single phase shifter and 12.5 dBV for a push-pull Mach-Zehnder Modulator. Such a modulator can be switched with a driving voltage of 3 $V_{pp}$ having insertion loss of 4 dB. In (b), the circles and crosses represent measured losses obtained from cut-back measurements [this work] and obtained from our previous work [35] using an OEO-material with a similar refractive index.

The voltage-length product decreases with wavelength and slot width and reaches values below 20 $V_{\mu m}$ for a slot width of 50 nm and a wavelength around 1.2 $\mu m$. In comparison, a value of 100 $V_{\mu m}$ is obtained at 1.55 $\mu m$. Thus, by taming the resonance of the OEO-material one can enhance the light-matter interaction by a factor of five. A similar performance enhancement can be obtained for all other slot widths. The simulated propagation losses are shown in Fig. 5(b). To confirm the validity of the simulated values we extract experimentally the propagation losses of 100 nm wide slots by means of cut-back measurements using a grating coupler setup (red circles). Additionally, propagation losses measured at a wavelength 1.6 $\mu m$ in MZMs using a similar OEO material (JRD1) are included as a reference, to verify the simulated propagation losses for other widths [35]. The propagation loss increases linearly for wavelengths larger than 1.2 $\mu m$ (plasmonic-dominated loss) and exponentially for smaller wavelengths (OEO material-dominated loss). Multiplying the curves in (a) and (b) leads to the loss-voltage-length product ($\alpha U_\pi L$), plotted in (c). This figure of merit decreases by a factor 2 when reducing the wavelength and reaches values of 25 dBV. The optimal point is reached when the OEO-material’s absorption starts to dominate the plasmonic losses which is the case at 1.2 $\mu m$ for a 50 nm-wide slot and 1.25 $\mu m$ for a 200 nm-wide slot. Please note that the stated voltage-length products are those of a phase shifter, while $U_\pi L$ reported in literature [13,31,49,52] mostly relate to a Mach-Zehnder modulator operated in push-pull configuration. This configuration allows to achieve a $\pi$-phase shift between both arms of the modulator by applying a $U_\pi$ on one arm and a - $U_\pi$ on the other arm in modulator with half the length. Loss-voltage-length products of 25 dBV (12.5 dBV) can then be reached for a simple phase shifter (push-pull Mach-Zehnder Modulator [24,31,43,44,46]). This means POH Mach-Zehnder modulators of 4$\mu m$ length can be switched from on-state to off-state by a driving voltage of ±1.5 V with insertion losses of 4 dB only. The electrical energy consumption then should not exceed a few fJ/bit. The loss-voltage-length product of POH modulators comes close to reach similar values as reported for metal-oxide-semiconductor (MOSCAP) of 7 dBV [32] or silicon-insulator-silicon capacitor (SISCAP) 13 dBV [33] optical modulators. However, the voltage-length product of POH-modulators (~0.04 $V_{\mu m}$) can be multiple times smaller compared to MOSCAP (~0.9 $V_{\mu m}$) or SISCAP (~1 $V_{\mu m}$) modulators.
The slot width dependency within these simulations suggests that narrowest slots are the preferred choice when the electro-optic coefficient is assumed to be independent from the slot width [31]. However, the electro-optic coefficient of current materials (JRD1, DLD-164, HD-BB-OH/YLD124 [this work – see experimental part]) drops for smaller slot-widths [35].

5. Experimental results - measuring the nonlinear enhancement

The previous section showed that POH modulators are best operated closer to the resonance frequency of the material. In this section we confirm the numerical expectations by experimental evidence, based on the measurement of the voltage-length product ($U/\lambda L$) [31,35]. The wavelength dependent $U/\lambda L$ product has been measured by using the experimental setup schematically depicted in Fig. 6

![Fig. 6. Experimental setup used to characterize the wavelength-dependent plasmonic phase modulator (PPM) performance. Two tunable lasers are alternatively used as a light source, covering the wavelength range from 1260 nm to 1370 nm and from 1460 nm to 1630 nm. Light is coupled to the chip by edge coupling. A sinusoidal 40 GHz RF-signal is generated and amplified before being applied via RF probes to the PPM. Finally, the modulated optical signal is measured with an optical spectrum analyzer to obtain the ratio between optical carrier and modulation sidebands.](image)

Two external-cavity tunable lasers are used as light sources to cover the wavelength range from 1260 nm to 1370 nm and 1460 nm to 1630 nm, respectively. Lensed fibers are used to couple light to and from the chip by edge coupling. A maximal optical power of 0 dBm was applied to the PPM and stable device operation was observed. No higher optical powers could be fed to the PPM due to the limited fiber to chip coupling efficiency. On the chip, light is guided by standard silicon waveguides (450 nm x 220 nm) buried in SiO$_2$. A sinusoidal 40 GHz electrical signal (blue line in Fig. 6) is generated by an RF synthesizer and amplified before being applied to the PPM via 67 GHz RF-probes. This frequency is well below the 3 dB bandwidth of POH modulators (>170 GHz) [44]. The modulated light is detected by an OSA to determine the voltage-length product based on the ratio between modulation sidebands and optical carrier [35]. The plasmonic phase shifters were fabricated in-house as discussed in [45]. The nonlinear material, a binary chromophore composite of 75 wt% HD-BB-OH and 25 wt% YLD124 was applied by spin coating after fabrication. Prior to the experiment, the devices have been poled by applying electrical fields of 180 V/μm to the plasmonic slot waveguides and heating the devices up to their glass transition temperature ($T_g$ $\sim$110°C), or slightly above, thus inducing the EO coefficient $r_{33}$ [45].

Figure 7 shows the measured voltage-length product ($U/\lambda L$) as a function of wavelength for devices having slot widths of 50 nm (a) and 200 nm (b). The blue dots represents the measured values.
Both cases show that $U_\pi L$ is up to three times lower at shorter wavelengths. The reduction can be traced back to the parameters $\lambda$, $n_{\text{EO}}$, $n_{\text{slow}}$ and $r_{33}$, see Eq. (3). These results agree well with the simulations from the previous section. From Eq. (3) we expect a four times smaller $U_\pi L$ for the device Fig. 7(a) with a 50 nm slot waveguide over the device Fig. 7(b) with a 200 nm wide slot waveguide as $U_\pi L$ is directly proportional to the slot width. However, only a 1.5 decrease can be observed. We attribute the discrepancy to a reduction of the electro-optic coefficient with the slot width [35]. The experimental electro-optic coefficient ($r_{33,\text{exp}}$) can be extracted from Fig. 7 and by:

$$r_{33,\text{exp}}(\lambda) = \frac{U_{\pi,\text{exp}}}{U_{\pi,\text{sim}}} \cdot r_{33,\text{exp}}(\lambda). \quad (5)$$

Here, $r_{33,\text{exp}}$ is the measured electro-optic coefficient while $r_{33,\text{sim}}$ is the electro-optic coefficient assumed in the simulation (180 pm/V). $U_{\pi,\text{exp}}$ and $U_{\pi,\text{sim}}$ are the experimentally and numerically determined $\pi$-voltages, respectively. The results are shown in Fig. 8 as a function of wavelength for a slot width of 50 nm (a) and 200 nm (b).

The dots represent measured values of the EO coefficient, while the dashed red line represents the theoretical wavelength dependence according to the two-state model [47]. $r_{33}$ reaches 90 pm/V and 325 pm/V for slot widths of 50 nm and 200 nm, respectively. We attribute the smaller electro-optic coefficient ($r_{33}$) for the 50 nm slot to a reduced poling efficiency, caused by three effects [35]. First, the poling efficiency depends on $w_{\text{slot}}$ due to surface effects. Second, the fabricated PPMs of this batch suffer from rougher waveguides than usual, causing an unstable poling process for slot widths below 80 nm. Previous batches show an unstable poling process only for slot widths below 40 nm. Third, a poling temperature above the glass transition temperature was applied during poling for the 50 nm slot. Considering the individual measurements of the 50 nm or 200 nm wide slots, we observe
that the trend line of $r_{33}$ follows the trend line predicted by the two state model quite well. The electro-optic coefficient enhances by 60% when reducing the wavelength from 1.6 μm to 1.25 μm. This can be observed for both cases, suggesting that the enhancement is independent of the slot width.

The performance of a modulator is ultimately given by the loss-voltage-length product. The wavelength dependency of this figure of merit is plotted in Fig. 9 for both slot widths. The blue dots are obtained from the measured voltage length product times the simulated propagation loss, while the dashed red line is solely obtained from simulation. The blue dots follow the simulated trend line, suggesting that for both slot widths the performance enhances by two times when the PPMs are operated in close vicinity to the OEO material’s resonance.

![Fig. 9. The normalized loss-voltage-length product plotted as a function of the wavelength for two different phase shifters (normalized with respect to the wavelength at 1.65 μm to make comparisons easier). It can be seen that the performance improves by up to a factor two at shorter wavelengths.](image)

6. Conclusion and outlook

We have demonstrated, for the first time, the potential to harness the resonance of active OEO materials to improve the device performance of practical modulators. Thereby, we have experimentally shown an enhancement of the modulator’s efficiency by almost 100% when operated within 200 nm of the OEO’s bandgap. It has been demonstrated that, while the enhancement is partly due to waveguide and plasmonic dispersion, the strongest contribution is provided by the dispersion of the electro-optic coefficient. We measured an in-device electro-optic coefficient ($r_{33}$) of 325 pm/V at 1.26 μm corresponding to a second order susceptibility ($\chi^{(2)}$) of almost 2000 pm/V. This is the highest in-device electro-optic coefficient achieved in plasmonic modulators to date.

Further, we have confirmed that the experimental results are in good agreement with our theoretical framework, which combines plasmonic mode analysis and the two-state model. This framework can be used to predict the performance of plasmonic waveguide structures and novel OEO-materials, as long as the electric-optic coefficient is known at one specific wavelength.

Finally, we would like to emphasize the unique prospects provided by plasmonics and their ability to tame material resonances. For instance, operating the MZM presented in [31] in close vicinity to the material’s bandgap could result in unprecedented $U_{π}L$ products of ~10 Vμm. Furthermore, the results are also of importance for chemists designing novel OEO-materials. Prior to this work, materials have been designed for a high electro-optic coefficient and low optical losses (dB/cm). Here, we have shown that the design of future OEO-materials is no longer subject to the optical losses as the imaginary part of the refractive index can be up to three orders of magnitude larger without increasing the overall losses. Thus material engineering can solely focus on maximizing the electro optic coefficient.

7. Appendix

7.1 Approximated figure of merit

The performance of plasmonic modulators is dominated by the losses ($\alpha$) which are given by
\[
\alpha_{\text{total}} = 10 \cdot \log_{10} \left( \exp \left( \frac{4\pi}{\lambda} \cdot n_{\text{eff}} \cdot L \right) \right) \quad (6)
\]

where, \(4\pi/\lambda n_{\text{eff}}^2\) is the attenuation coefficient and \(L\) the length of this modulator. The requirement of a phase modulators is that the accumulated phase shift after the length \(L\) equals \(\pi\):

\[
\pi = \Delta \beta \cdot L = \frac{2\pi}{\lambda_0} \cdot \Delta n_{\text{eff}} \cdot L \quad (7)
\]

Substituting (7) into (6) and keeping in mind that losses are minimal when the argument of the exponential term is minimal leads to the following term for the efficiency \((\eta)\) which should be maximized:

\[
\eta = \frac{\Delta n_{\text{eff}}}{n_{\text{eff}}} \quad (8)
\]

The following approximations are used for the qualitative discussion in Section (3). First, the change in the refractive index is proportional to \(n_{\text{OEO}}^3 \cdot r_{33}\). Second, the losses \((n_{\text{eff}}^\prime)\) are caused by the imaginary part of the refractive index of the metal or of the OEO-material and are approximated by \(n_{\text{eff}}^\prime = n_{\text{OEO}}^\prime + n_{\text{Metal}}^\prime\). Thus the efficiency is:

\[
\eta = \frac{\Delta n_{\text{eff}}}{n_{\text{eff}}} = \frac{n_{\text{OEO}}^3 \cdot r_{33}}{(n_{\text{OEO}} + n_{\text{Metal}})} \quad (9)
\]

### 7.2 Refractive index of the OEO material

| Wavelength [nm] | \(n^\prime\) | \(n^\prime\prime\) |
|-----------------|-------------|-----------------|
| 1200            | 1.875       | 0.0047          |
| 1250            | 1.844       | 0.0019          |
| 1300            | 1.822       | 0.0008          |
| 1310            | 1.819       | 0.0007          |
| 1350            | 1.806       | 0.0004          |
| 1400            | 1.795       | 0.0002          |
| 1450            | 1.784       | 0.0001          |
| 1500            | 1.776       | <0.0001         |
| 1550            | 1.770       | <0.0001         |
| 1600            | 1.765       | <0.0001         |
| 1650            | 1.760       | <0.0001         |

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