New paradigms in materials and devices for hybrid electro-optics and optical rectification

Lewis E. Johnson,[1,2,*] Delwin L. Elder,[1,2] Huajun Xu,[2] Scott R. Hammond,[1,2] Stephanie J. Benight,[1] Kevin O'Malley,[1] Bruce H. Robinson,[2] Larry R. Dalton[2]

1. Nonlinear Materials Corporation, Seattle WA 98109
2. University of Washington Department of Chemistry, Seattle WA 98195

*Corresponding author: lewisj@uw.edu or lewisj@nonlinearmaterials.com

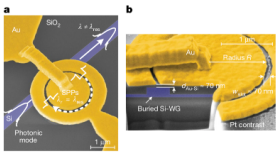
ABSTRACT

We review recent transformative advances in materials design, synthesis, and processing as well as device engineering for the utilization of organic materials in hybrid electro-optic (EO) and optical rectification (OR) technologies relevant to telecommunications, sensing, and computing. End-to-end (from molecules to systems) modeling methods utilizing multi-scale computation and theory permit prediction of the performance of novel materials in nanoscale device architectures including those involving plasmonic phenomena and architectures in which interfacial effects play a dominant role. Both EO and OR phenomenon require acentric organization of constituent active molecules. The incumbent methodology for achieving such organization is electric field poling, where chromophore shape, dipole moment, and conformational flexibility play dominant roles. Optimized chromophore design and control of the poling process has already led to record-setting advances in electro-optic performance, e.g., voltage-length performance of < 50 volt-micrometer, bandwidths > 500 GHz, and energy efficiency < 70 attojoule/bit. They have also led to increased thermal stability, low insertion loss and high signal quality (BER and SFDR). However, the limits of poling in the smallest nanophotonic devices—in which extraordinary optical field densities can be achieved—has stimulated development of alternatives based on covalent coupling of modern high-performance chromophores into ordered nanostructures. Covalent coupling enables higher performance, greater scalability, and greater stability and is especially suited for the latest nanoscale architectures. Recent developments in materials also facilitate a new technology—transparent photodetection based on optical rectification. OR does not involve electronic excitation, as is the case with conventional photodiodes, and as such represents a novel detection mechanism with a greatly reduced noise floor. OR already dominates at THz frequencies and recent advances will enable superior performance at GHz frequencies as well.

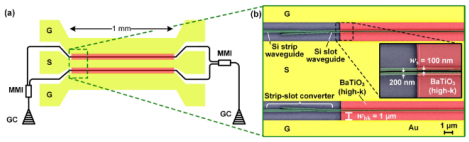
INTRODUCTION

Organic electro-optic (OEO) materials have seen a recent renaissance due to hybrid device architectures, which maximally leverage their strong Pockels effect response in nanophotonic devices with strong optical mode confinement and with fabrication methods compatible with state-of-the art photonics processes. Such platforms include silicon-organic hybrid (SOH),[1-4] in which OEO materials are integrated with silicon photonics, plasmonic-organic hybrid,[1, 4-6] (POH) which use plasmonic structures to further enhance field confinement, and architectures involving III-V semiconductors such as indium phosphide.[7] These platforms have attracted recent commercial interest due to meeting a need for high-bandwidth, compact, power-efficient modulators for photonic integrated circuits (PICs), with applications[8-13] ranging from optical interconnects/datacom to microwave photonics, mmWave telecom technologies, satellite communication, metrology/test systems, and defense-related technologies.

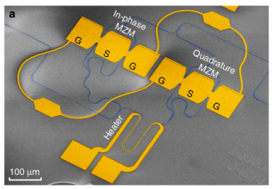
The last few years have seen a variety of performance breakthroughs from hybrid OEO technologies, including > 500 GHz bandwidth in a Mach-Zehnder interferometer (MZI),[14] < 100 aJ/bit power consumption in a high-speed IQ modulator,[15] modulator footprints[16] < 20 um^2, monolithic integration with high-speed BiCMOS electronics and stable operation under required thermal conditions,[17] and compact, low-loss, high-performance modulators implemented on silicon photonics platforms.[18, 19] A selection of recent devices are shown in Figure 1.



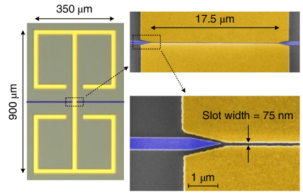
High-efficiency microscale POH ring resonator (Haffner *et. al. Science* **2017**)



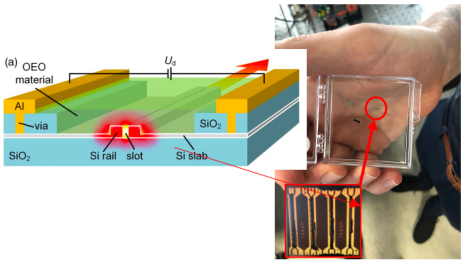
Ultrafast capacitively coupled SOH MZI (Ummethala *et. al. Optica* **2021**)



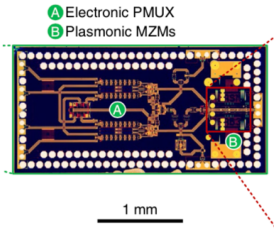
sub-100 aJ/bit IQ modulator (Heni *et. al. Nature Comm.* **2019**)



60 GHz, sub-mm plasmonic mixer/antenna (Salamin *et. al. Nature Photonics* **2018**)



Ultra-low voltage, sub-mm SOH MZI (Kieninger *et. al. Optica* **2018**)



Integrated POH-BiCMOS transmitter (Koch *et. al. Nature Electronics* **2020**)

Figure 1. A selection of recent breakthrough hybrid OEO devices, including an ultra-compact POH ring resonator (upper left), > 76 GHz capacitively coupled SOH MZI (upper center), an ultra-efficient high-speed POH IQ modulator (upper right), a compact mmWave POH mixer (lower left), a low-voltage sub-mm SOH MZI (lower center), and an a > 100 Gbit/second integrated POH-SiGe BiCMOS transmitter (lower right).

Present hybrid OEO approaches require electric-field poling of the OEO materials to induce acentric ordering of the active chromophores and obtain a Pockels effect response, a process that declines in efficiency as waveguide widths become smaller[20, 21] and that requires optimization of an additional manufacturing step. While poling provides a feasible route for the next generation of devices, and current hybrid OEO approaches are already showing significant advantages as a technology for augmenting the performance of active components in silicon photonics,[1, 10, 22, 23] intrinsically ordered OEO materials suitable for current nanophotonic device architectures would simplify wafer-scale fabrication and enable even greater densities of active components. Present hybrid approaches also rely on high-speed photodetectors, which require additional heterogeneous integration or coupling and lose efficiency at high sub-THz frequencies. Pockels effect materials such as OEO materials also exhibit a complementary effect, optical rectification (OR),[24] which can be used as an alternative photodetection technique that does not require electronic excitation and enables efficient detection at higher frequencies. While previously insufficiently efficient to compete with state-of-the-art photodetectors, the combination of increases in OEO material performance and optical mode confinement greatly increases the viability of OR as a detection technique and the ability to use organic NLO materials in multiple categories of active components.

The key figure of merit for electro-optic phase modulation is $V_\pi L$, or the product of voltage and length required for a phase shifter to induce a half-wave phase shift. In a MZI configuration, a half-wave phase shift yields complete destructive interference, enabling digital switching. $V_\pi L$ is a property of both material performance ($n_e^3 r_{33}$) and mode confinement (w/Γ), as illustrated in Figure 2, which also details the contribution to r_{33} from materials properties, including the contributions from hyperpolarizability and acentric order. The state-of-the art $V_\pi L$ values for SOH (320-500 V-µm) and POH platforms (50-100 V-µm) are compared with conventional free carrier dispersion (PN junction) based silicon photonics and thin-film lithium niobate (TFLN)[25] in Figure 2, illustrating the potential for much smaller devices at CMOS-level drive voltages without the need for low-noise amplifiers (LNAs). Contributing factors to the performance of these platforms are discussed in subsequent sections.

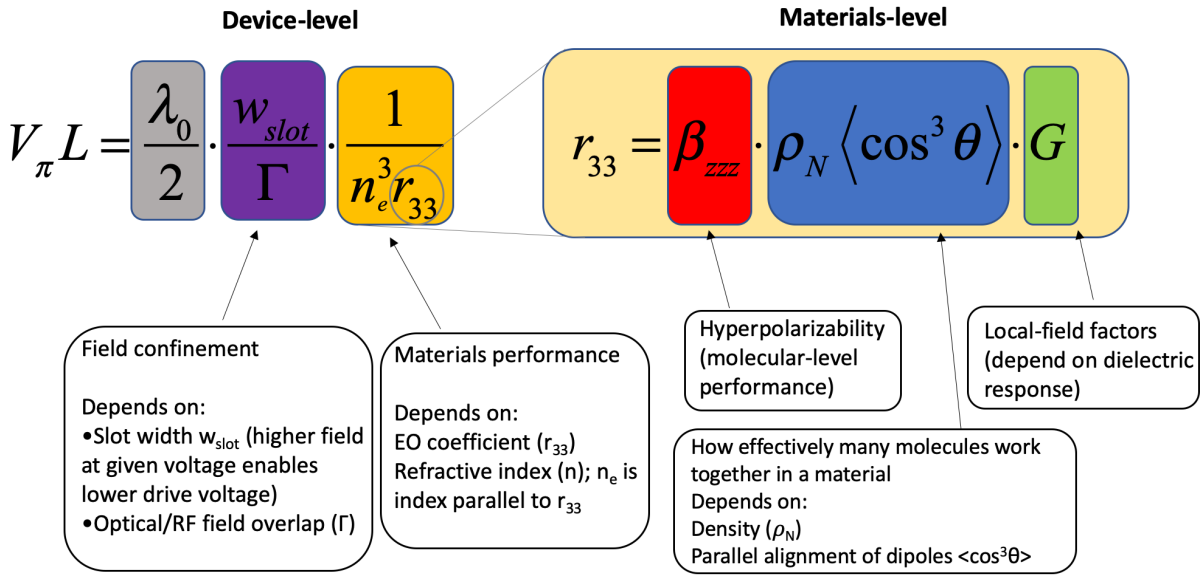


Figure 2. The voltage-length product of an electro-optic device represents the phase shifter length L_π required at a drive voltage V_π required to achieve a half-wavelength phase shift in light passing through the phase shifter. It is a function of the confinement of optical and electrical/RF fields in the waveguide and of the electro-optic performance of the active material. In a Pockels effect device, this is quantified by the electro-optic coefficient r_{33}, which is a function of individual chromophore nonlinearity, concentration, and ordering.

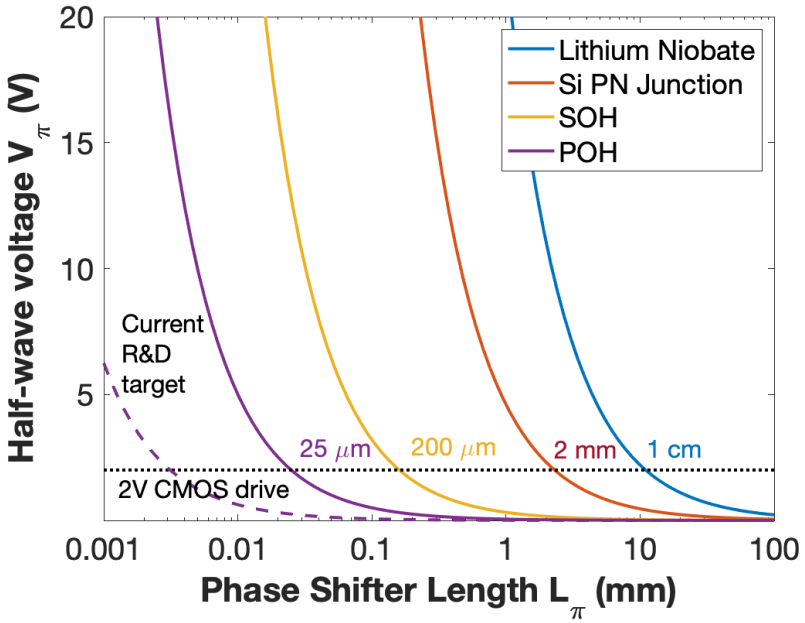


Figure 3. $V_\pi L$ frontiers, showing the phase shifter length L_π required for digital switching at 2V drive for various technologies. Current SOH devices have a $V_\pi L$ of about 400 V-μm[3, 18] and current POH devices about 50 V-um.[20] Present R&D goals are targeting an 8-10x reduction in these values due to a mixture of chromophore improvement and improving ordering in narrow waveguides to better leverage confinement effects.

CHROMOPHORE DEVELOPMENT

Recent development work on OEO chromophores[1, 26-31] has focused on increasing r_{33} (and reducing $V_{\pi}L$) via increased hyperpolarizability (β) under the constraints of acceptable optical loss and processibility for efficient poling, as well as increasing thermal stability of these high-performance materials. We have conducted a large-scale theory-guided, quantitative structure-property relationship-based design effort to increase EO activity, which has led to new records in hyperpolarizability[30] and to electro-optic activity in excess of 1000 pm/V.[31] Such activity represents the first time that an OEO material has exceeded the performance of barium titanate ($BaTiO_4$), without the large mismatch between optical and RF dielectric properties[32] that greatly complicates design of devices using $BaTiO_4$ and similar materials, particularly at high frequencies. Such a screening effort for OEO materials also focuses time-consuming synthesis efforts on prospects with the best predicted properties. Figure 4 compares a large computational screen with a set of well-characterized prior chromophores and highlights prospects that have been synthesized. These prospects have generally tracked (and exceeded)[30, 31] computational predictions of their performance. Over 16 new chromophore designs and variants have been synthesized and characterized.

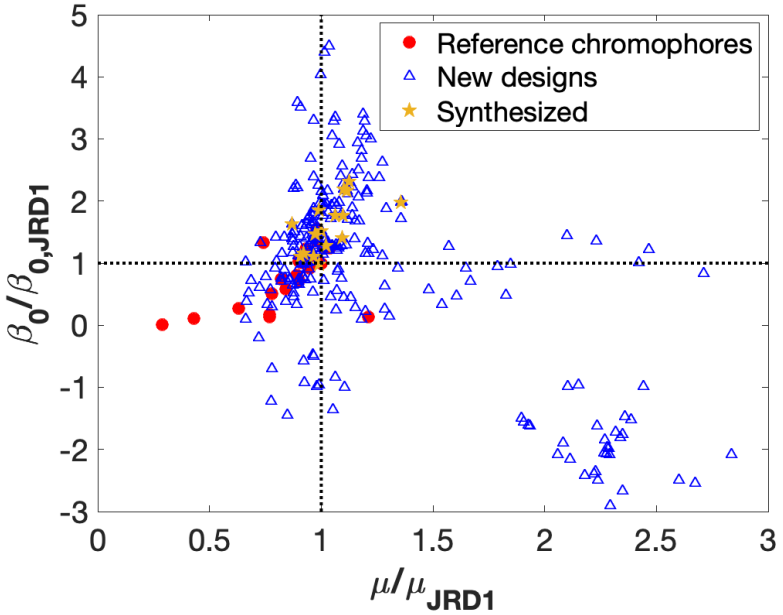


Figure 4. Computed (M062X/6-31+G(d) in PCM chloroform) hyperpolarizability as a function of ground-state dipole moment, showing 266 candidate chromophores and 25 reference chromophores. 16 of these new designs (plus variants with equivalent truncated representations) have been synthesized since 2017.

The high molecular hyperpolarizability of these chromophores is combined with optimized side-chains for poling as neat materials, avoiding the need for a polymer host and maximizing ρ_N.[33-35] When combined with appropriate charge-blocking layers[31, 35] to minimize leak-through current during poling, the materials designed through this process have enabled a two-fold increase in bulk electro-optic activity since 2017, as shown in Figure 5. These materials are only now beginning to be tested in high-performance nanophotonic devices. As processing protocols are improved, we expect to see commensurate increases in in-device r_{33} and reduction in $V_{\pi}L$, likely enabling POH devices with performance < 20 V-μm with poled materials, even without substantial further improvement in acentric order.

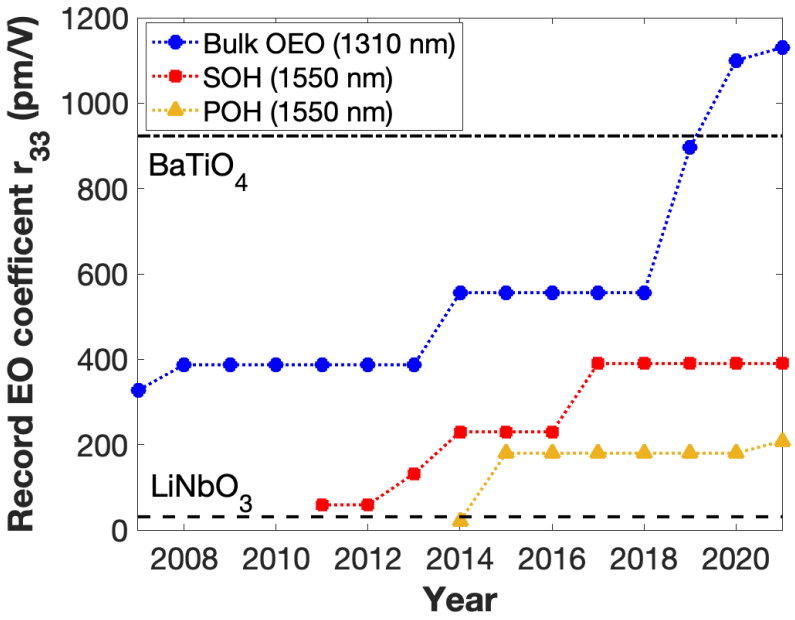


Figure 5. Record r_{33} for bulk (thin film) and nanophotonic devices as a function of time, showing the recent increase in bulk performance due to theory-guided design of high-β chromophores as well as the 2-3 year time lag in translating materials performance to nanophotonic device performance.

PROCESSING ADVANCES AND STABILITY

Additional areas of focus of R&D efforts on OEO materials for hybrid architectures are thermal and photochemical stability of high-r_{33} materials. Thermal stability can be greatly improved via crosslinking,[36, 37] a technique that has historically been applied to polymer systems but recently applied to neat chromophore systems such as HLD,[29] in which two complementary chromophore monomers, HLD1 and HLD2, undergo a thermally-induced reaction to form a durable thermoset plastic, with the glass transition temperature T_g increasing by ~100°C during the crosslinking process. Structures for HLD1 and HLD2 and their parent chromophore are shown in Figure 6.

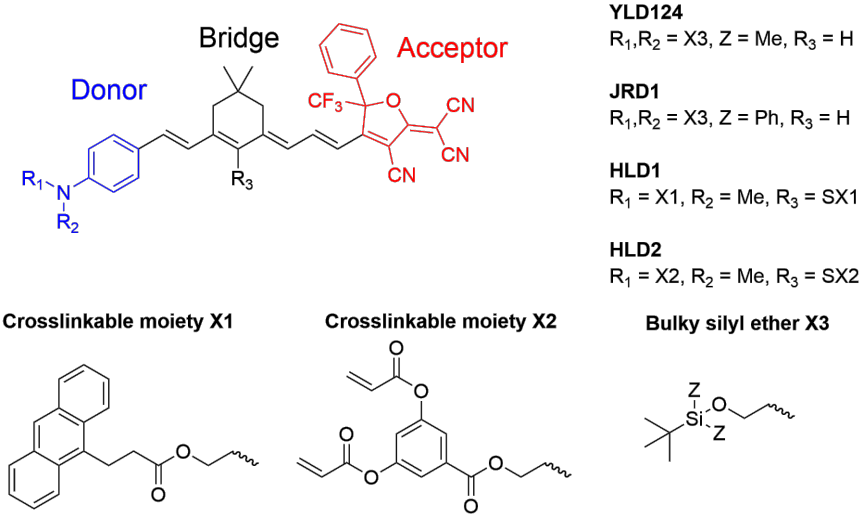


Figure 6. Commercially available high-performance chromophores based on evolved CLD-type architectures. The entire family of chromophores shares an arylamine electron donating moiety, ring-locked tetraene bridge, and CF_3-phenyl substituted tricyanovinylfuran electron acceptor.[36]

The crosslinking process for HLD enables high EO activity (up to 300 pm/V at 1310 nm) to be preserved while stabilizing the material to be able to withstand extended operation at high-temperature. In addition to in-device operation,[17] long-term storage stability has been demonstrated for thin-film HLD EO devices at temperatures of 85°C, 105°C, and 120°C. Devices were poled in a thin-film configuration on 20 nm PVD TiO_2 charge blocking layers, crosslinked to > 150 °C, with temperature increased at 10° C every 10 minutes after poling temperature was reached. Devices were stored in N_2-purged ovens (emulating hermetic packaging) with n=7 devices in the 85°C condition, n = 6 devices in the 105°C condition, and n = 5 devices in the 120°C condition. Environmental stability testing is underway to define packaging requirements. A summary of stability data is shown in Figure 7; stable long-term performance was maintained under all three conditions.

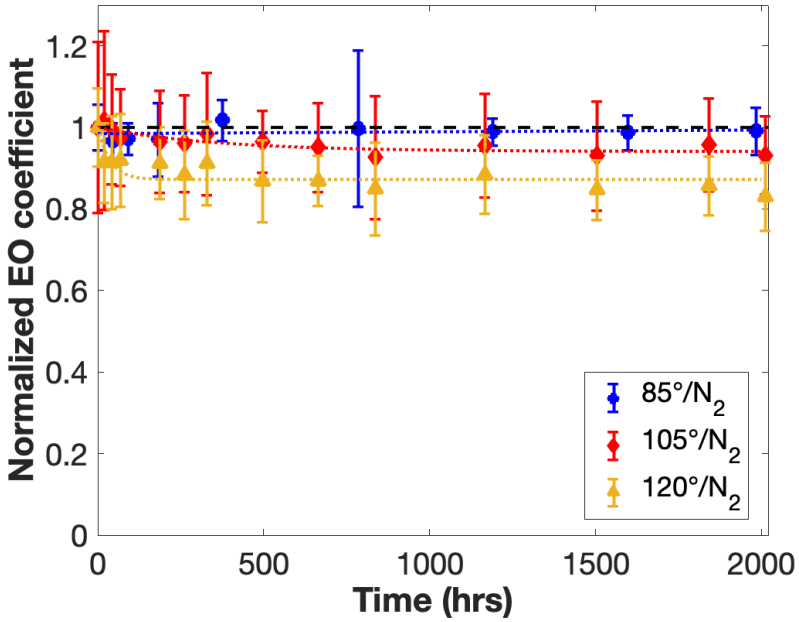


Figure 7. Thermal stability of EO activity of crosslinked thin films of 2:1 HLD1:HLD2 over 2000 hours of high-temperature exposure in an inert atmosphere. Dotted lines are least-squares fits – linear for 85°C (negligible slope) and exponential burn-in to an equilibrium value (105°C and 120°C). After initial burn-in, performance levels off with 94% retained after 2000 hrs at 105°C and 87% at after 2000 hrs at 120°C.

SEQUENTIAL SYNTHESIS

While impressive EO performance and thermal stability have been obtained with poled materials, direct synthesis of ordered multilayers of chromophores presents the potential for even higher performance and thermal stability. In sequential synthesis, a monolayer of chromophores is deposited on a surface, with additional layers added through solution processing via removal of protecting groups and coupling of complementary groups in a manner analogous to peptide synthesis, with optional lateral coupling to increase stability. Such an approach[36] was pioneered by Marks[38] and co-workers with earlier-generation chromophores prior to the development of modern hybrid device architectures, but could not produce sufficiently thick films for the all-polymer devices being developed at the time. If applied to nanophotonic devices, where only 10-25 layers may be required to form sufficiently thick films, a sequential synthesis approach could be used to increase acentric order from $\langle \cos^3 \theta \rangle$ of ~0.2 to ~0.8 with high thermal stability, and incorporate current-generation chromophores to leverage their increased hyperpolarizability and achieve exceptional electro-optic activity. This approach is particularly suited to recently demonstrated planar POH architectures,[39] but could be accomplished in other architectures via careful selection of materials and coupling chemistries. The sequential synthesis processes and some potential chemistries are schematically illustrated in Figure 8.

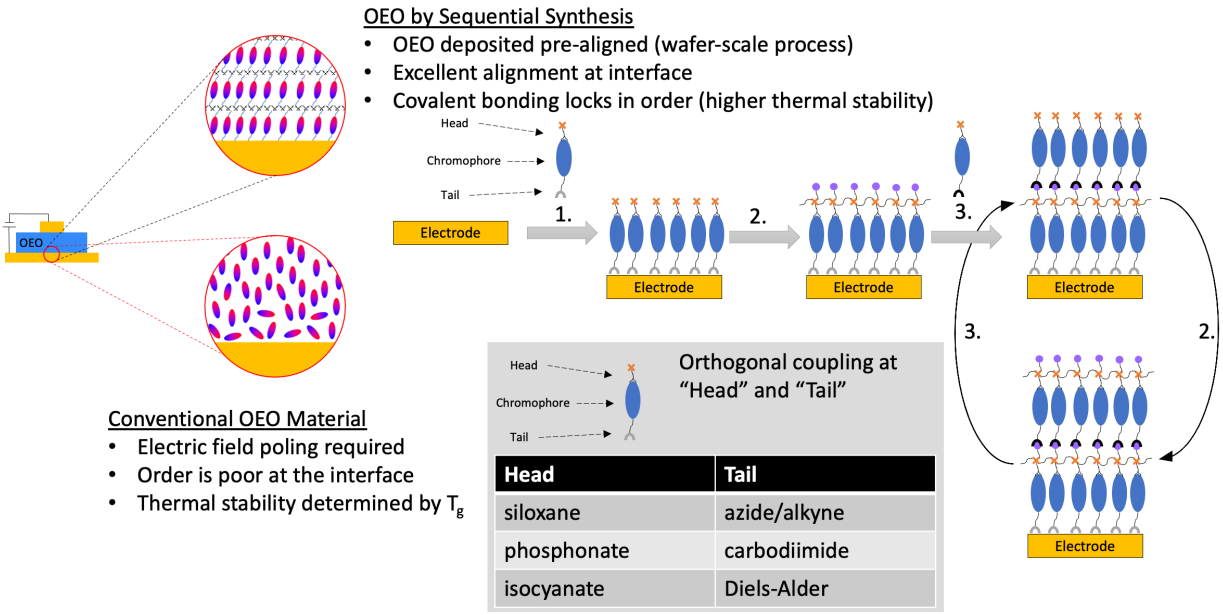


Figure 8. Schematic of sequential synthesis of an aligned chromophore film on an electrode using complementary head and tail functional groups and comparison with electric-field poling

OPTICAL RECTIFICATION

Optical rectification in OEO materials on silicon was first observed over 16 years ago,[24] with the potential for high performance at frequencies in the hundreds of gigahertz modeled several years later, suggesting the potential for > 9 mA/V at 400 GHz assuming a 300 pm/V OEO material, a refractive index of 1.7, and a slot width of 100 nm.[40] This would suggest that with a refractive index of 1.85, a 1000 pm/V OEO material, and a 50 nm POH device with a Γ of 0.7, responsivity could exceed 1.8 A/W applying the same scaling illustrated in Figure 2. Such a device has not yet been demonstrated, but would have a responsivity exceeding current photodetection technologies with the potential for very low noise.

CONCLUSION AND OUTLOOK

Over the past four years, hybrid OEO devices have demonstrated exceptional performance in bandwidth, power efficiency, and space efficiency, becoming a technology of significant interest[10, 22, 23] for improving the performance of active components in silicon photonics platforms. We have demonstrated recent increases in materials performance through theory-aided design, as well as high thermal stability and the ability of OEO materials to survive demanding conditions if devices are appropriately packaged. The high performance of new materials may enable optical rectification as a next-generation photodetection technology and both performance and manufacturability can be further improved via sequential synthesis replacing electric field poling as a method for inducing acentric order.

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FINANCIAL INTEREST DISCLOSURE

Drs. Johnson, Elder, Hammond, and O'Malley are employees of Nonlinear Materials Corporation, which provides materials and services related to organic electro-optic materials discussed in this work. Dr. Benight is a consultant in lieu of staff to Nonlinear Materials Corporation. Prof. Robinson is an advisor to Nonlinear Materials Corporation. Drs. Johnson, Elder, Hammond, O'Malley, and Benight and Prof. Robinson hold equity positions in Nonlinear Materials Corporation. Dr. Xu and Prof. Dalton declare no competing financial interests.

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