Polymer Silicon Hybrid Systems: A Platform for Practical Nonlinear Optics†

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Nonlinear optical polymers can provide a unique and powerful platform for building a number of useful optical devices. Traditionally, they have been used in all-polymer waveguiding systems, which have had a number of severe practical limitations. Silicon has recently become a popular material for integrated optical systems, but it lacks strong nonlinear properties. By combining nonlinear optical polymer claddings and silicon waveguides in a hybrid system, we show that a number of significant advantages are obtained. In particular, fabrication can be done using conventional CMOS processing, with the polymer added as part of back-end processes (after all of the high-temperature processing is completed), and the waveguide structure can also serve as an ultracompact set of electrodes. We discuss some optical devices that have recently been built into this system, which already show superior performance to anything achieved in silicon or polymer alone. We also propose using second-order nonlinear polymers with silicon for parametric amplification and oscillation and predict that such devices could exhibit extraordinary performance.

1. Introduction

Recently, a number of advances in both design and synthesis of nonlinear optical polymer material systems have been achieved. Polymers with a second-order optical nonlinearity, with r_{33} values of 200 pm/V,[1] have been demonstrated. This r_{33} is around an order of magnitude larger than that of many conventional nonlinear optical materials, like lithium niobate, which has an r_{33} of 30 pm/V.[2] Third-order nonlinear optical polymers have also been synthesized, with Kerr coefficients of 7×10^{-14} cm^2/W,[3] around twice that of silicon.[4] At the same time, great advances have been made in the field of silicon photonics; low-loss waveguides, high-Q resonators, and electrically pumped all-silicon modulators have been built into chip-scale systems.[5,6]

Nonlinear optical polymers and silicon waveguides can be combined to provide a hybrid platform with unique advantages. The nonlinear polymer can provide a flexible source of large second- and third-order optical nonlinearities. At the same time, the silicon can provide tight optical confinement due to its high index of refraction and optically invisible electrical contacts to the active regions. Moreover, fabrication technology for silicon is quite advanced due to the significant investments made by the silicon microelectronics industry. It is likely that a silicon−polymer hybrid system will prove to be an attractive platform for a number of optical devices, including integrated optical parametric oscillators, broad-band optical amplifiers, low-V_π modulators, and all-optical switches.

2. Results and Discussion

2.1. Silicon Photonics. The low-loss properties of crystalline silicon in the near-infrared have been known for decades.[7] In the last six years, in particular, the field of silicon photonics has witnessed explosive growth. For linear optics, silicon waveguides have proven extremely versatile, in that advanced lithographic and processing techniques have enabled feature sizes as small as 40 nm to be realized[8] and Q values of 300k to be obtained.[9] This has been achieved, moreover, in planar, lithographic processes that are compatible with commercial silicon foundries, suggesting that manufacturing costs in quantity will be very modest. Modulation at speeds up to 10 GBit/sec has also been demonstrated on the basis of the free-carrier plasma dispersion effect, with V_π values of 1.8 V obtained.[10] Surprisingly, even all-silicon photodetection at high speeds is possible through the introduction of specifically tailored lattice defects.[11]

Silicon-based nonlinear optics is a very young field. This is largely due to the absence of an appreciable second-order nonlinearity due to inversion symmetry in the silicon lattice. Attempts have been made to strain the silicon layer in order to achieve second-order nonlinearities,[12] but these experiments have not resulted in a large r_{33} value nor any practical devices to date. While parametric gain has been achieved through third-order optical nonlinearities in silicon, the requisite pump powers are extremely large, around 11 W.[13] These high power levels, which are well in excess of the damage threshold of silicon waveguides, require that devices be operated in pulsed mode and greatly limit the devices' utility. The picture is similar for Kerr-effect-based all-optical switches; the amount of gate optical power needed for π radians of phase shift, which we will refer to as P_π, is typically on the order of 100 W.[14] While lower switching powers have been obtained through the use of two-photon absorption (TPA) to generate free carriers, these devices are not ultrafast, with bandwidth limitations of around 1 GHz or less.[15]

2.2. Nonlinear-Polymer-Based Modulators. The most familiar application of nonlinear optical polymers is electrooptic modulation.[1,16] Nonlinear polymer modulators are based on the Pockels effect,[2] in which an electric field changes the refractive index of a poled material, generally for near-infrared radiation. Mach−Zehnder modulators and ring modulators can both be built on this principle. Waveguides are generally built by performing photolithography on a series of different polymer materials, creating a multilayer, low index contrast guiding system. Nonlinear-polymer-based devices have conventionally

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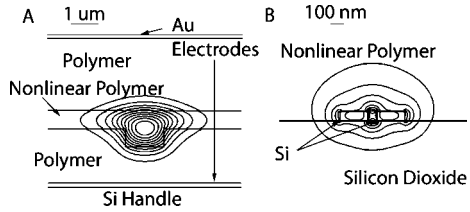

Figure 1. Panel A shows the diagram of a nonlinear polymer waveguide, fabricated in an all-polymer system. Three layers of polymer are used; the middle layer is a nonlinear polymer that will shift in response to an electric field. The modal pattern for the fundamental TM mode near 1.55 um is also shown; contours of |Ey| are plotted. Electrodes are provided by the two gold layers; a field can be induced vertically and thus shift the effective index of the waveguide. Panel B shows a modulator built with similar polymers but in a silicon system; in this case, a slot waveguide is used to provide both a waveguide and a set of electrodes. The modal pattern for the fundamental TE mode near 1.55 um is shown, and contours in |*E*| are drawn.

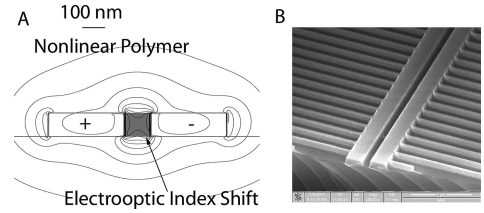

Figure 2. Panel A shows a diagram of a slot waveguide with a contour plot of the TE optical mode near 1.55 um. The region in which the electrooptic index shift occurs due to charging the two silicon arms is highlighted. Panel B shows a SEM micrograph of a cleaved slot waveguide. The tiny arms that extend normally from the waveguide allow electrical contact with the two waveguide arms but are optically invisible. Note that the charges indicated on the arms are due to an applied voltage and do not imply any difference in doping concentrations; typical designs that we have used involve uniformly doped near-intrinsic silicon.

been built using mutlilayer photolithographic processes in all-polymer systems with metal electrodes. Figure 1 shows a diagram of waveguides suited for use in electrooptic modulators.[16]

Conventional nonlinear polymer modulators are already quite competitive with modulators based on other material systems. One of the most important characteristics of an electrooptic modulator is the V_π, or level of external voltage that is needed to induce π radians of phase shift in a Mach−Zehnder. This corresponds to switching from complete optical transmission to complete extinction. A lower value is desirable because it makes the supporting electronics cheaper and vastly reduces the amount of power that is consumed by the modulator; the on-chip power dissipation for electrooptic modulators is an important limitation toward achieving massive integration.[10] With nonlinear polymers, V_π values of 0.65 V have been demonstrated. For comparison, modulators based on lithium niobate, a nonlinear optical crystal, have only achieved V_π values ofaround 2 V to date.[18] Lithium niobate devices are the most common Mach−Zehnder modulators available commercially today.

Silicon-based modulators have also not reached the levels of performance exhibited by nonlinear polymer modulators, with V_π values of only 1.8 V achieved.[10] Green et al.[10] have recently shown that a very low $V_\pi L$ value can be obtained if one offsets the switching signal with a DC bias, suggesting that improvements to this might be possible; however, these designs are predicated on a large amount of current flowing through the device, suggesting that even if lower switching voltages are obtained, power usage may remain unacceptably high. By contrast, very little current flows through a nonlinear polymer modulator, as the nonlinear polymers usually have low conductivities.

When integrated with a silicon platform, however, polymer-based modulators achieve even greater advantages.[17] Devices are fabricated by defining a series of waveguides and optical structures on a silicon-on-insulator substrate. The polymer can then be spin coated to obtain a high-quality film. This has the advantage of requiring high-resolution lithography to be done only with silicon rather than polymer. The main device complexity in such an approach is captured in the silicon layer, which is fabricated using standard CMOS processing steps.

The integrated nonlinear polymer−silicon design involves a slot waveguide, which consists of two distinct silicon strips. As shown in Figure 1, a TE optical mode is supported that will be concentrated largely in the slot area. This concentration is due to the sharp discontinuity from the silicon, with a refractive index near 3.48, to the polymer, with typical refractive indices from 1.5 to 1.7. Because the polymer is not very conductive, both halves of the slot waveguide can be held at different potentials. Figure 2 shows a closer view of the optical mode in the slotted waveguide,[8] with the region that would exhibit index shift due to different arm voltages highlighted. The DC field too is concentrated between the slot waveguide, precisely where the optical mode is localized. The entire voltage drop of the modulating signal then occurs over a length that can be only 0.1 μm or less. Compared to a more conventional modulator, where the voltage drop might occur over the course of 9 μm or more,[16] as would be the case for the all-polymer design shown in Figure 1, a modulator based on this design will clearly have a much lower driving voltage, even for otherwise equivalent polymers.

It is important to note that the silicon in this case is being used only as an optically transparent conductor and, as such, is uniformly doped near the optically active regions. Our work in the past[17,19] has involved lightly doped p-type silicon with 10^{15} boron/cm^3. The only significance of the doping level is that it decreases the resistivity compared to that of intrinsic silicon and can cause increased optical loss in high quantities.

Slot waveguides have been fabricated with low optical losses through electron beam lithography; losses of better than −4 dB/cm have recently been obtained.[8] Slot waveguide modulators including polymers have also been built; ring-based modulators have been built, which exhibit tuning of 5.2 GHz/V, around five times more responsive than modulators designed with similar polymers on a nonslotted architecture. When used in a Mach−Zehnder configuration, we have recently obtained $V_\pi L$ values of around 0.5 V·cm; this suggests that V_π values below 250 mV could be obtained in a device of several cm of length in a push−pull configuration without any improvement in material performance.[19] A recent design study suggests that it should eventually be possible, by using the best nonlinear polymer materials available today and scaling down the slot size of the silicon guides, to obtain further improvements; slot-waveguide-based modulators with V_π values of 10 mV or less should be possible, an improvement over current typical V_π values by nearly 2 orders of magnitude.[20]

2.3. Nonlinear Polymer All-Optical Modulators. Recently, all-optical modulators built in silicon have been the subject of increased interest. Such devices are usually built based on a Mach−Zehnder geometry; a signal optical mode goes through both arms, and a gate optical mode is introduced into one of the arms. As illustrated in Figure 3, a nonlinear refractive index

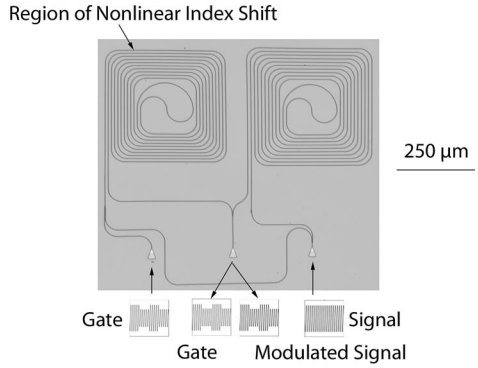

Figure 3. Diagram of a nonlinear all-optical modulator. The optical micrograph is of a typical realization in a silicon ridge waveguide for radiation near wavelength 1.55 μm. The spooled arms are approximately 1 cm in length.

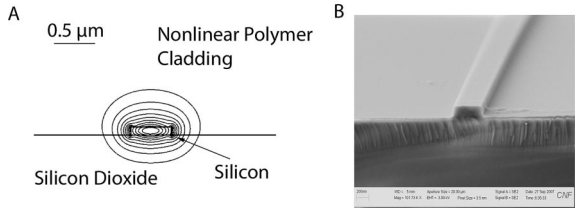

Figure 4. Panel A shows a diagram of the polymer clad ridge waveguide, with a contour plot of the fundamental TE mode. Panel B shows a SEM image of a cleaved waveguide.

shift in one of the arms can then lead to all-optical modulation, whereby the gate optical mode can switch the signal optical mode. The gate and signal optical modes can be at any frequencies that are guided by the waveguides, as long as they are far enough apart to be distinguished. In most realizations, the nonlinear polymer would be deposited on both arms of the all-optical modulator; nonlinear cross talk would only occur in one waveguide since the gate optical mode only propagates in one arm. Such devices could someday form the basis for efficient all-optical wavelength conversion or even the basis for an all-optical logic system. Depending on the nonlinear mechanism, the operating bandwidth can be as high as the terahertz.[21] The particular device shown in Figure 3 relies on a 0.5 × 0.1 μm silicon ridge waveguide, which is shown in Figure 4.

By analogy to the V_π value reported for the electrooptic modulator, we can characterize the response strength of an all-optical modulator in terms of P_π, the instantaneous power required to obtain an induced π radians of phase shift in one arm. Lower values for P_π are better as they correspond to more responsive modulators that require less gate power. Nearly all optical waveguides have a certain amount of optical power that can be tolerated (a damage threshold); higher values than this lead to device breakdown, often due to the effects of heating. Achieving lower P_π values is an important step toward practical all-optical modulators since it is extremely desirable to build devices that can function with continuous-wave optical inputs.

One approach to lowering the P_π is to use a nonlinear material with a stronger response. Nonlinear polymers show great promise in this area; ultrafast nonlinear coefficients double that of silicon have already been demonstrated.[3] In fact, polymers with around 10 to 100 times the third-order nonlinear coefficient of silicon are likely to soon be demonstrated.[22]

In addition to selecting a stronger material, the nonlinear response of a device can be enhanced by concentrating the optical modes into a smaller propagation area. In such a situation, a larger electric field will be induced by the optical mode, which will, in turn, lead to a larger shift in the nonlinear material. The amount of all-optical modulation obtained in a waveguide can be written as

$$\Delta\varphi = k_0\left(\frac{3}{4}\chi^3\frac{Z_0}{n^2}\right)\frac{P}{A_{\text{eff}}}L$$
$$A_{\text{eff}} = \frac{Z_0^2\left(\int 2\text{Re}(E\times H^*)\text{d}A\right)^2}{4n^2\int E_x^4 + E_y^4 + E_z^4\text{d}A} \quad (1)$$

Here, A_{eff} is the effective area for the optical modes involved, the E and H fields are for the modal patterns, and L is the length. In a standard fiber optic cable with a mode field diameter on the order of 10 μm, the effective area would be approximately 100 μm^2. However, a ridge silicon waveguide with dimensions of 0.5 × 0.1 μm, such as that shown in Figure 4, will have an effective area for a nonlinear cladding close to 0.2 μm^2; this implies that for identical nonlinear materials and optical powers, the phase shift due to all-optical modulation would be increased by a factor of 500 for a given interaction length.

Utilizing this silicon−polymer hybrid geometry, we have previously demonstrated an all-optical modulator with terahertz bandwidth. A P_π of 2 W was obtained.[21] By contrast, in all-silicon devices, typical values were obtained for an ultrafast P_π range from 50 to 100 W.[14,23] As nonlinear polymers continue to increase in strength, they will be easily integrated into the polymer−silicon hybrid system used for this device. It is likely that P_π values of less than 1 W will eventually be possible, even without resonant enhancement. Our hope is that these devices can cross into the regime of exhibiting signal gain, where a small amount of power on a gate can be used to modulate another higher amount of power on a signal wavelength. Such a device is the basic building block of an ultrafast all-optical logic system.

2.4. Nonlinear Polymer Optical Amplifiers and Parametric Oscillators. One of the outstanding goals in silicon photonics has been the development of optical amplifiers and lasers for the near-infrared. Due to silicon's indirect band gap, it has not been possible to demonstrate either amplification or lasing with electrical pumping. A further difficulty for the near-infrared is that silicon's band gap of 1.12 eV is larger than the photon energy near 1550 nm, which is 0.8 eV.[24] However, optically pumped gain elements can still be built. The strongest effect in silicon that can be used to this end is stimulated Raman scattering. Gain coefficients of 0.6 W^{-1} cm^{-1} have been achieved, leading to 1 dB/cm of gain with a pump beam of around 400 mW. This has, in turn, led to the demonstration of optical amplifiers[25] and silicon Raman lasers.[26] Some work has been done with optical parametric amplification based on silicon's χ3 moment, but this effect is weaker than the Raman effect, and only pulsed operation has been achieved.[13]

There are several severe limitations with the Raman effect; first, the optical pump and the amplified optical beam must be separated by a frequency characteristic of the material. In silicon, this frequency separation is 15.6 THz.[27] This implies that to amplify a signal near 1550 nm, one would require a pump at 1434.3 nm, but to amplify a signal near 1580 nm, one would require a pump at 1460 nm. Moreover, the Raman gain bandwidth is only around 100 GHz,[6] suggesting that a Raman-based amplifier could only amplify a limited set of optical inputs. For the same reasons, a Raman laser could only be tuned through a limited frequency regime without changing pump wavelengths.

A number of advantages can be obtained by using a nonlinear polymer cladding with a silicon waveguide system. The most

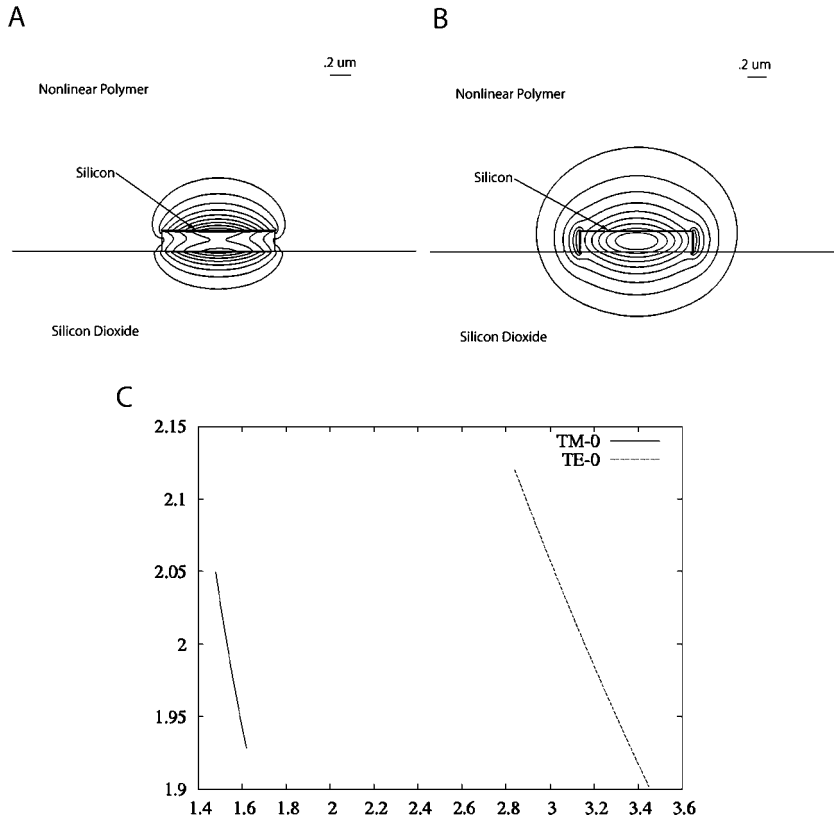

Figure 5. Panel A shows the modal pattern of the fundamental TM mode of a 0.2 × 0.6 μm silicon ridge waveguide, while panel B shows the modal pattern of the fundamental TE mode of the same waveguide. In both cases, contours are plotted of $|E|$. The dispersion diagram is shown in panel C. Phase matching could be achieved using this waveguide for a pump near 1.5 μm and a signal and idler near 3 μm.

important benefit is the additional nonlinearity available from the polymer as compared to the nonlinearity from silicon alone. The $\chi 2$ moment that can be obtained from nonlinear polymers is quite large; an r_{33} of 200 pm/V[1] corresponds to a $\chi 2$ moment of around 8.3×10^{-10} m/V. The induced nonlinear polarization moment for a 1×10^7 V/m electric field, typical for the 0.5 × 0.1 μm silicon ridge waveguides described here is then 8.3×10^4 V/m. By contrast, the $\chi 3$ moment of silicon is 1.7×10^{-19} m^2 V^{-2}, corresponding to an induced nonlinear polarization of only 170 V/m.[4,28] This analysis presumes, of course, that the r_{33} encountered for electrooptic modulation is, in fact, an ultrafast effect. The polymer response bandwidth has been shown with conventional modulators to be at least 165 GHz.[29] Through optical rectification, however, we have established that at least some nonlinear polymers do show a relatively flat r_{33} value to even the optical frequency.[17]

Equation 2 predicts the gain coefficient available for a waveguide with a second-order nonlinearity[2]

$$g = \chi^2 \varepsilon_0 4 \left(\frac{P_{\text{pump}}}{A_{\text{eff}}} w_{\text{signal}} w_{\text{idler}} \right)^{1/2} \frac{1}{(2n)^{3/2}} Z_0^{3/2}$$
$$A_{\text{eff}} = \frac{Z_0^3}{(2n)^3} \left(\frac{\left(\int 2\text{Re}(\mathbf{E} \times \mathbf{H}^*) \cdot \mathbf{z} \text{d}A \right)^{3/2}}{\int E^*_{\text{signal}} E_{\text{pump}} E^*_{\text{idler}} \text{d}A} \right)^2 \quad (2)$$

As for the third-order nonlinearity, a smaller effective area leads to a larger nonlinear effect and, in this case, higher gain. Typical effective areas for a second-order nonlinear process in a ridge silicon waveguide will be around 0.5 μm^2. In this case, the approximate gain coefficient for parametric gain in a $\chi 2$ nonlinear polymer with signal and idler near 3 μm would be 104 cm^{-1} $W^{-1/2}$ for a polymer with r_{33} of 200 pm/V, implying that a gain of 1 dB/cm could be obtained with only 10 μW of optical power in the pump beam. Note that the gain coefficient in the case of a second-order nonlinearity depends on the square root of the optical power, in contrast to the Raman gain coefficient reported previously.

Clearly, a large amount of optical gain can potentially be obtained with a far lower power pump beam through the use of nonlinear polymers than can be obtained with the Raman effect. This could lead to optical amplifiers with very low pump power. However, another significant advantage is the intrinsically broadband nature of the nonlinear polymer response. Unlike the Raman effect, no resonance is involved with the parametric gain process in a $\chi 2$ polymer. As a result, in principle, a single pump wavelength could be used to provide optical gain to signals at a wide array of frequencies. Similar to a conventional material with optical gain, when the gain exceeds the loss in a polymer system, spontaneous emission is then amplified, and coherent output can be created through the use of a resonator. With a nonlinear optical system, such a device is known as an optical parametric oscillator (OPO).[2]

The threshold of such a device depends heavily on the waveguide loss that is present. The lowest threshold Raman lasers have been built into systems with waveguide losses of 0.2 dB/cm and achieve a threshold of 26 mW.[26] If losses of 5 dB/cm were obtained with a polymer−silicon waveguide system with the previously calculated gain, then the threshold for a nonlinear polymer-based OPO with an r_{33} of 200 pm/V could be around 130 μW, even if we assume no substantial resonant enhancement of the pump. Of course, lowering the waveguide losses toward the level obtained in the silicon system would lower the threshold power as well.

One challenge with the nonlinear polymer approach is that one must satisfy the well-known equations for parametric gain[2] given in eq 3; the frequency and the wavenumber of the signal to be amplified, as well as a secondary "idler" beam, must both add to the pump frequency and wavenumber

$$w_{\text{pump}} = w_{\text{signal}} + w_{\text{idler}}$$
$$k_{\text{pump}} = k_{\text{signal}} + k_{\text{idler}} \quad (3)$$

For a $\chi 2$ process, eq 3 precludes having all three frequencies spectrally near one another. However, it is possible to build a nanoscale silicon waveguide which will support a number of different optical modes, at significantly different frequencies. Silicon will not show substantial losses even at wavelengths as long as 100 μm, except for a small region near 10 μm.[30] The main difficulty of eq 3 is that the effective indices of all of the involved wavelengths must be matched. This should be possible, however, in a number of ways. First, the high index contrast nature of silicon makes it possible to engineer situations in which different polarizations have very different effective indices. For example, a 0.2 × 0.6 μm silicon ridge waveguide would have a TM mode near 1.5 μm as well as a TE mode near 3 μm, which would have nearly identical effective indices. Figure 5 shows a diagram of this waveguide and the field pattern. Due to the high index contrast of the waveguide, the different polarizations will still have some nonlinear cross talk for any choice of poling direction, though maximizing this value is a significant design challenge.

Once the polymer is deposited on a silicon waveguide such as the one proposed in Figure 5, it would need to be poled. This could be achieved by charging the waveguide by a fairly sparse series of contacts, as well as depositing an external electrode layer, perhaps on top of the polymer with a metal layer. Another possibility would be a second silicon strip on the side of the waveguide, forming a lateral voltage drop. The best solution will depend, in part, on what direction is optimal for the second-order nonlinearity, which will, in turn, depend on the precise wavelengths and waveguide geometries under consideration. The chip would then be heated and a voltage maintained by means of these two electrical paths, which would establish the orientation of the polymer. We have used this approach to poling successfully in the past.[17]

Another possibility is to pole the polymer periodically;[31] since the polymer is not grown in the fashion of a nonlinear optical crystal but instead deposited on the silicon and then oriented via an induced electric field, this could be a practical solution. A periodically poled nonlinear material would be able to compensate for an arbitrary wavenumber mismatch in eq 3, though it would then require that the silicon waveguide have a more extensive set of electrical contacts. One way to achieve this would be through the use of segmented waveguides.[8]

A silicon and $\chi 2$ polymer system could perform a number of useful functions on the basis of the high gain coefficient observed. Broad-band amplification could be obtained in the telecom wavelengths, perhaps with a pump near 1200 nm and an idler near 5 μm. Another possibility is the generation of a frequency in the mid-infrared with an OPO. Tunable, continuous-wave sources are not widely available in this regime, and those that are available are extremely expensive.[32] As a result, the silicon−polymer system has the potential to act as the basis of a new class of inexpensive, widely tunable, room-temperature optical sources in this frequency regime.

3. Conclusions

It is clear that the silicon−polymer system offers a series of significant advantages over both silicon and polymer optical structures, combining the best of both worlds. From the silicon, we get the ability to build nanoscale features, to rapidly scale complexity, and to massively concentrate optical and electric fields. From organic nonlinear materials, we gain access to extremely high optical nonlinearities and the ability to engineer material properties for specific applications. This opens up a path to fundamentally new on-chip optical devices, including parametric oscillators, modulators with extraordinary performance characteristics, and all-optical logic elements exhibiting signal gain. Perhaps the most exciting feature of this material system, however, is its adaptability; as new soft materials emerge, it will be possible to integrate them very rapidly into the silicon platform, providing fundamentally new functionality. In the future, this may include stronger nonlinearities, saturable absorption, and perhaps electrically pumped light emitters.

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