1 Introduction

1.1 Motivation

It has become increasingly recognized [1] that "photonic integration" is an important next step in the evolution of computing, telecommunications, sensing, transportation, medical, defense, and entertainment technologies. Such integration permits the best features of electronics and photonics to be exploited for information technology applications. There are important technological drivers of photonic/electronic integration, including realization of improved bandwidth and thermal management – transporting and manipulating information using photons avoids the high-frequency resistive losses and heating associated with movement of electrons in metal. Photonic/electronic integration is also evolving to include "chipscale" integration wherein both electronic and photonic circuitries are integrated on to the same chip, analogous to complementary metal-oxide semiconductor (CMOS) electronic integration, which has revolutionized computing. Potential advantages of chipscale integration include far-reaching improvements in size, weight, power consumption, performance, reliability, and cost. While chipscale photonic/electronic integration is not likely to be monolithic, as in CMOS electronic integration, and the problems to be faced will certainly be challenging, there can be little doubt that it will ultimately occur and will have considerable societal and economic impact. Such integration has been greatly advanced by recent developments in the field of silicon photonics, plasmonics, and metamaterial device architectures [2 to 21]. For example, the high index of refraction of silicon has permitted a striking reduction in the size of photonic circuits, making the dimensions of these circuits more compatible with chipscale integration. Further reductions in circuit dimensions appear possible by exploiting plasmonics.

A key component of photonic/electronic integration is the interconversion of signals between the electronic and photonic domains. This is where electro-optics comes into play. An electro-optic (EO) material is one in which the electrical fields associated with photons and electrons can communicate through a highly hyperpolarizable (easily perturbed by electric fields) charge distribution. For an electro-optic material to be optimum for the transduction of electronic signal information into photonic signal information, the charge distribution should be easily perturbed by small electric field potentials (ideally by millivolt electric field potentials to minimize power consumption) and should have very fast (ideally femtosecond) response to time-varying electrical fields. It is these features that have attracted attention to organic EO materials.

The fundamental response time of conjugated π-electron systems is the phase relaxation time and is almost universally of the order of tens of femtoseconds. Moreover, conjugated π-electron molecular systems exhibit exceptional hyperpolarizability, suggesting that a few millivolts applied to devices may be used to encode electronic information onto optical transmissions and to manipulate that information (e.g., route or steer, wavelength division multiplex or color code).

Unlike inorganic crystalline EO materials, which are typically naturally occurring, many organic (π-electron) EO materials are created by design, and a virtually endless stream of new materials can be created. This seemingly endless ability to create new organic EO materials is both an incredible opportunity and challenge. The opportunity lies in ever-increasing electro-optic activity (and thus reduced power requirements) and in realizing special properties such as conformal and flexible materials, materials that can be effectively integrated with a wide variety of disparate materials (metals, metal oxides, semiconductors, inorganic glasses, etc.), and materials amenable to a wide variety of processing options, including low-cost manufacture of complex device structures by techniques such as soft and nano-imprint lithography. The downside of the seemingly endless structural modifications possible with organic EO materials is the difficulty of deciding which materials to focus upon for device applications.

Two decades of defining structure/function relationships critical for device applications have already occurred. This effort has led to marked improvement of some properties. For example, during the first decade of the twenty-first century, the electro-optic activity of organic materials was improved at nearly a Moore's Law rate, while auxiliary properties were improved to satisfy Telcordia standards. Indeed, it is now likely that organic electro-optic materials yield the fastest response times (bandwidth performance to 15 THz has been demonstrated in difference-frequency generation [22], in all-optical modulation [5], and in pulsed studies [23]) and the largest electro-optic effect (∼500 pm/V compared with ∼30 pm/V for the commercial standard lithium niobate, $LiNbO_3$). With silicon–organic hybrid (SOH) devices, digital signal processing bandwidths greater than 100 Gbit/s with power consumption of less than 1 femtojoule/bit have been demonstrated. Thus, the commercial potential of organic electro-optics for next-generation information technology is becoming increasingly recognized. Electro-optic devices based on organic materials sold by Gigoptix have satisfied Telcordia requirements. Still further improvement in performance is likely required for chipscale integration applications and for certain other device applications such as spatial light modulation. This book provides important insight into how necessary improvements are likely to be achieved and what will be the ultimate limits to performance of EO materials.

1.2 Overarching objective of this book

The purpose of this book is to provide clear insight into the structure/function relationships critical to optimizing the performance of devices based on organic electro-optic materials. This explicitly includes those relationships critical for optimizing

electro-optic activity, optical loss, stability, etc. The lessons learned from the past 20 years of research are reviewed, and suggestions are made as to how these lessons can be used to further advance the performance of organic electro-optic materials. An important objective of this book is to provide critical understanding of state-of-the-art theoretical methods that have been employed to guide the selection of nonlinear optical chromophore structures for large molecular first hyperpolarizability, and to nano-engineer component structures to achieve the non-centrosymmetric organization of chromophores necessary for large macroscopic electro-optic activity. An introduction to techniques for characterizing all of the relevant properties of electro-optic materials is also important, as these techniques provide the data tables desired by engineers utilizing materials for device and systems applications. Characterization data are also critical components of the feedback loop necessary for optimizing material performance. A unique feature of organic electro-optic materials is their exceptional processability, and this book attempts to provide insight into this diversity of processing methodologies. Finally, organic electro-optic materials provide an impressive array of new application possibilities, and an understanding of current and emerging device concepts is thus an important objective of this book.

This book is aimed at providing advanced undergraduate students, graduate students, postgraduate students and faculty, together with industrial scientists and engineers, with a comprehensive understanding of the field of organic electro-optics. A particular objective is to provide a knowledge base that facilitates individuals seeking to enter the field of organic electro-optics either as researchers or applications engineers. To that end, the book commences with an overview of the central concepts of nonlinear optics.

The book can also be an important reference text for those desiring to understand nano-engineering of materials dependent on spatially anisotropic interactions. It should also prove helpful to those desiring to learn the theoretical methods, such as Monte Carlo and molecular dynamics methods, used to simulate the assembly of individual molecules into macroscopically ordered lattices. Electro-optic materials are attractive model systems for understanding forces that dominate macroscopic order, since non-centrosymmetric (acentric) symmetry is required for finite electro-optic activity for materials based on dipolar chromophores. Study of electro-optic activity provides critical insight into which theoretical methods are most effective in designing improved materials and in understanding order/disorder processes associated with various processing protocols. The insights gained from considering organic electro-optic materials can be invaluable aids to improving organic electronic, photovoltaic, photorefractive, and light-emitting materials. In general, this book can serve as a primer for the systematic nano-engineering of soft-matter electroactive materials.

1.3 Overview of topics covered

Chapter 1 focuses on the perspective and motivation of this text and provides a brief introduction to topics covered and to a brief history of organic electro-optics. Although discovery of the Pockel's effect (electro-optics) pre-dates the laser, the introduction of

the laser clearly paved the way for nonlinear optics because of the large electric fields associated with intense laser beams. The advent of fiber optic telecommunications opened avenues for commercial application of electro-optic materials. Chapter 1 provides a brief general introduction to fundamental phenomena of nonlinear optics, which are discussed in Chapters 2 and 3. Chapter 2 provides an important introduction to the terminology of nonlinear optics, which is necessary for understanding the subsequent chapters. Figures-of-merit introduced in this chapter are particularly important for engineers attempting to consider electro-optic materials for device and systems applications and in guiding the choice among different material options. This chapter also provides an introduction to the measurement techniques used in the characterization of various macroscopic optical nonlinearities and related physical properties. Chapter 3 continues in the spirit of Chapter 2, but narrows the focus to electro-optic phenomena. Chapter 4 considers molecular hyperpolarizabilities, including theoretical methods of computing and experimental techniques for measuring such hyperpolarizabilities. This chapter illustrates the fundamental approaches to the design of organic electro-optic molecules (chromophores). Chapter 5 deals with self-assembled materials; Chapter 6 deals with crystalline materials; and Chapter 7 focuses on poled polymers (with a brief section on thermo-optic polymers). These three chapters cover the three fundamental approaches to generation of macroscopic electro-optic materials. Chapter 8 provides an overview of applications while Chapter 9 considers specifics of device structures. Chapter 9 provides a more detailed discussion of various stripline and resonant electro-optic device structures. Chapter 10 focuses on other second-order nonlinear optical effects including frequency doubling, optical rectification, and difference-frequency generation. Chapter 11 deals with photorefractivity, which is an important application of the electro-optic effect. Chapter 12 provides conclusions and future prognosis, particularly in light of emerging competing technologies.

1.4 A brief history

The Pockel's effect [24] pre-dates the discovery of the laser; however, it was the discovery of the laser that opened the field of nonlinear optics and promoted identification and definition of various second-order (electro-optic modulation, second-harmonic generation, optical rectification, difference-frequency generation, etc.) and third-order (phase conjugation and four-wave mixing, third-harmonic generation, all-optical modulation, etc.) nonlinear optical effects. For his seminal contributions to nonlinear optics, Nicolaas Bloembergen was awarded the Nobel Prize in Physics in 1981 and is viewed as the father of nonlinear optics [25 to 27].

It is difficult to provide a meaningful history of organic nonlinear optics, as many hundreds of exceptional scientists have contributed tens of thousands of important papers contributing to the evolution and definition of the field. Thus, any brief history must be incomplete and to a great extent arbitrary in coverage. With that caveat, we attempt to provide a brief perspective on events and circumstances that led to the current state-of-the-art. Perhaps the first motivation for consideration of organic π-electron

systems for nonlinear optics was provided by French theorists [28 to 32] who suggested that the long conjugation lengths of π-electron systems could give rise to large optical nonlinearities. Indeed, it was suggested that first molecular hyperpolarizabilities might increase with a fourth power dependence on π-conjugation length and that second molecular hyperpolarizabilities might increase with a sixth power dependence on conjugation length. Of course, early calculations were overly simplistic, and the combination of electron Coulomb and electron–phonon interactions resulted in limitations on delocalization and thus on the increase in optical nonlinearity with increasing π-conjugation (chromophore) length. Moreover, the charge transfer absorption band of chromophores undergoes a bathochromic solvatochromic shift with increasing length, and thus optical loss becomes a serious problem even at telecommunication wavelengths for the longest π-electron molecules (chromophores). This revision of expectations regarding organic electro-optic materials based upon improved understanding of the complexity of these materials was to become the hallmark of organic electro-optic materials research over the next two decades.

Major advancement in fields of science usually requires substantial financial investment, and in the late 1980s such an investment was made by international companies and by the US government in the area of nonlinear optics. In the United States, a major stimulus was provided by the Defense Advanced Projects Research Agency (DARPA) in a program managed by Dr. John Neff that involved Hoechst-Celanese Corporation, Lockheed Martin Corporation, and others, as well as a number of universities. A major motivation for DARPA interest was the potential for high-bandwidth time-division-multiplexing (TDM) possible with the ultrafast response times of π-electrons to time-varying electrical fields. By the late 1980s and early 1990s, outstanding research teams at Eastman Kodak, DuPont, IBM, 3M, Hochest-Celanese, Lockheed Martin, Boeing, Allied Signal, Eni Chem, Akzo, NTT, Battelle, British Telecom, etc. were producing organic electro-optic materials by electric field poling, sequential synthesis/self-assembly, and crystal growth. Crystals such as methylnitroaniline (MNA) and poled polymers based on incorporating stilbene and azobenzene chromophores into commercially available polymer hosts were extensively studied. The computation of molecular hyperpolarizabilities was rapidly advanced by groups such as those led by Anthony Garito (Univ. of Pennsylvania), Paras Prasad (SUNY-Buffalo), Brian Pierce (Hughes Aircraft Company), Tobin Marks and Mark Ratner (Northwestern), and many groups in Europe and Asia [33 to 36]. Auxiliary properties such as thermal stability were being extensively studied by researchers in Europe and the US (see Chapter 7). Individuals such as Seth Marder and Joseph Perry (then at the Jet Propulsion Laboratory/Cal Tech) popularized organic nonlinear optical materials with the chemistry community by describing structure/function relationships in terms familiar to organic chemists [35]. Proceedings of the SPIE (International Society of Optics and Photonics) and the Materials Research Society provide good snapshots of the remarkable evolution of organic nonlinear optics from the mid-1980s to the mid-1990s.

And then the bottom dropped out in the mid-1990s. Most of the industrial efforts were shut down, and funding from US Federal agencies was limited to small grants. One notable exception was Lockheed Martin Corporation, which continued research on

chromophore/polymer composite materials and contributed greatly to the understanding of optical loss in these materials [37,38]. The reasons for this sudden "market crash" of organic electro-optic materials and device research are likely three-fold: (1) The driving force for interest in organic electro-optics in the late 1980s likely was related to anticipated need for high (TDM) bandwidth in telecommunications, and it was realized that organic materials afforded the potential for high bandwidth as well as large optical nonlinearity and novel processing options. By the early 1990s it was realized that techniques such as wavelength-division-multiplexing (WDM) and code-division-multiplexing (CDM) could provide much of the needed bandwidth. (2) Corporations realized that organic electro-optics did not provide the market potential initially anticipated and particularly not at the level of core corporate activities focused on organic fibers (e.g., DuPont) and liquid crystalline materials (e.g., Hoehsct-Celanese). (3) By the mid-1990s, the electro-optic activity of organic materials still had not exceeded that of lithium niobate, and questions about the thermal and photochemical stability of organic materials had risen to the point of high concern. Moreover, the production of organic electro-optic materials was seen to have little in common with traditional polymer processing. In short, the engineering of transformative organic electro-optic materials was turning out to be more difficult than anticipated and the market potential not as great as anticipated.

It should be kept in mind that the evolution of organic electro-optics occurred at a time when polyacetylene (discovered in the 1980s) was being considered as having great promise for battery applications. Moreover, organic light-emitting device (OLED), photorefractive, and photovoltaic materials were attracting great interest. Enthusiasm for each of these types of organic electroactive materials waxed and waned during the late 1980s and early 1990s. It is only at the present time that commercial application has become a reality for OLEDs, and the greatest potential still lies in the future for each of these technologies.

Fortunately, a few research (mostly academic) groups in Europe, Asia, and North America remained active in research related to organic electro-optic materials in the late 1990s, and during this period several events occurred that were to produce a Moore's Law rate of improvement in the performance of organic electro-optic materials in the first decade of the twenty-first century. One of the events was the introduction of the tricyanovinyl furan (TCF) acceptor into chromophore synthesis [39 to 41]. This acceptor permitted an improvement in both molecular first hyperpolarizability and photochemical stability. The introduction of the TCF acceptor was also accompanied by introduction of isophorone group(s) into polyene bridges to improve the photochemical stability of this very effective bridge moiety. For poled polymer materials, an even greater advance occurred with the utilization of multi-scale modeling (coupled quantum and statistical mechanical methods) to optimize the efficiency of electric field poling. In the late 1980s, only 1–4% of molecular optical nonlinearity (first hyperpolarizability) was being effectively translated to macroscopic electro-optic activity because of deleterious effects of chromophore–chromophore dipolar interactions. An article in *Science* in 2000 marked the turning point in use of theory-guided improvement in electrically poled organic electro-optic materials [42]. With these advances in chromophore design

and processing, the electro-optic activity of organic materials exceeded that of lithium niobate and a wide range of prototype devices were successfully demonstrated.

The development of other photonic materials has certainly impacted the commercial potential of organic electro-optic materials. The development of low-loss silica fiber transformed telecommunications and stimulated the development of modulated lasers and electro-optic devices as methods of encoding electronic information onto long-range transmissions. As already noted in this chapter, the evolution of silicon photonics and demonstration of hybrid organic electro-optic/silicon photonic devices is having a significant effect, with promise of an even greater future impact, particularly on the chipscale integration of electronics and photonics.

New issues in material processing and device performance have been identified in the preceding decade, including the role that buffer layers play in controlling conductivity in organic electro-optic materials. Unlike organic electronic, photovoltaic, and light-emitting device materials, conductivity is best avoided in electro-optic materials. Interface engineering has become an important activity for all cases of organic electroactive materials. The introduction of thin interfacial layers of materials such as titanium dioxide to control charge injection and extraction has become commonplace in the fabrication of certain electro-optic devices.

Substantial progress has been made in the theoretical modeling of nonlinear optical effects (see Chapters 4 and 7), including the increased use of Møller–Plesset (MP) and density functional theory (DFT) quantum computational approaches for estimation of molecular hyperpolarizabilities and the dependence of hyperpolarizabilities on dielectric permittivity and optical frequency. Monte Carlo and molecular dynamics statistical mechanical methods have provided considerable insight into the role of various electrostatic interactions in influencing chromophore organization needed for macroscopic second-order optical nonlinearity. Recently, coarse-graining of statistical mechanical computations has permitted quantitative simulation to be extended to large systems.

Some additional seminal texts and reviews are cited in references [43 to 71], and more references are available in the subsequent chapters.

1.5 Units and conversion factors

A certain amount of confusion exists with respect to definitions (conventions) and units, and we address these in various chapters in this book. To start with, it has been common practice to report values for molecular hyperpolarizabilities in terms of centimeter-gram-second or cgs units (that is, in terms of electrostatic units or esu) while electro-optic coefficients are most commonly reported in SI (meter-kilogram-second or MKS) units (picometers/volt). To maintain connectivity with the substantial literature, we shall continue this duality. The conversion factors employed to go between SI and cgs units depend on the convention used for defining polarization in SI (or MKS) units. In the first convention, which we denote MKS(1), $P^{(n)} = \varepsilon_0 \chi^{(n)} E^{(n)}$ where $P^{(n)}$ is the polarization, ε_0 is the vacuum dielectric constant (= 8.8542×10^{-12} coulomb^2/Nm^2), $\chi^{(n)}$ is the nth-order optical susceptibility, and $E^{(n)}$ is the electric field. In the second convention,

which we denote MKS(2), $P^{(n)} = \chi^{(n)}E^{(n)}$. This is also the convention relevant to cgs units. Thus, $(4\pi\varepsilon_0)(10^{-4}c)^{1-n}\chi^{(n)}_{cgs} = \varepsilon_0\chi^{(n)}_{MKS(1)} = \chi^{(n)}_{MKS(2)}$ where c is the speed of light (= 2.9979 × 10^8 m/s). With appropriate attention to conventions used, $\chi^{(2)}$ in cgs units can be related to $\chi^{(2)}$ in MKS(1) units, i.e., 1 statvolt (stV)/centimeter (cm) = 4.1888 × 10^{-4} volt (V)/meter (m). There is also confusion in terms of the conventions employed in the definition of the nonlinear susceptibility and molecular hyperpolarizability expressions appearing in power- or Taylor-series expansions of polarization in terms of electric field. The reader is referred to subsequent chapters for in-depth discussions of various conventions and conversion factors, or to Ref. [72].

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