

Optical properties of nonlinear optical polymers: a method for calculation

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Methods for calculating the optical properties as a function of wavelength for second-order nonlinear optical polymers are presented. By combining the optical properties of the individual components, that is, those of the polymer and those of the chromophore, one can determine the real and the imaginary parts of the index of refraction and the second hyperpolarizability as a function of wavelength for a composite polymer, for either a guest-host or a side-chain system. These properties are important for characterizing and applying nonlinear optical polymers to devices. The methods allow for an accuracy in index of 3%, if the wavelength range includes the main absorption band (with absorption maximum at λ_{max}) of the chromophore, and of 0.2% for wavelengths 400 nm and more to the long-wavelength side of λ_{max} . In addition, if the index of the composite polymer is known at one specific wavelength on the long-wavelength side of λ_{max} , the dispersion in the index in this wavelength range can be predicted to within 0.1%. © 1998 Optical Society of America

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1. INTRODUCTION

Second-order nonlinear optical (NLO) polymers are organic polymeric molecules to which a NLO chromophore is either added as a guest or chemically attached in the main chain or as a pendant side chain. These chromophores are designed and synthesized to produce large second-order optical nonlinearities and to have thermal stability for application in optoelectronic devices. Clearly, if one has prepared a NLO polymeric film of good optical quality, then it is obvious that one would measure the index, absorption, and hyperpolarizability directly. However, to synthesize many different chromophores, substitute them into polymers, and cast them into thin films for their characterization is expensive and time consuming. One would, therefore, like to predict the optical properties when designing NLO devices without involved chemistry, film formation, and various methods of characterization. We outline procedures that show convenient and accurate ways to do this calculation.

The overall second-order nonlinearity $\chi^{(2)}$ depends on the molecular first-order hyperpolarizability β , the extent of orientation $\langle \cos^2 \theta \rangle$, the concentration of the chromophore in the polymer, and local electric-field factors. Although the concentration is usually low ($\sim 20\%$), its presence makes a significant contribution to the optical properties of the composite system. A number of these NLO chromophores have been studied, their β values and wavelengths of maximum absorption, λ_{max} , have been tabulated,^{1–5} and several second-order NLO polymers have been reviewed by Miller.²

In this paper we show that, by knowing the properties of the individual components (dye, neat polymer), one can

usually determine combinations with good accuracy. For the neat polymer the relationship between the index and the wavelength is given by a Sellmeier equation. To calculate the index of the dye from its absorption bands, both the Lorentz equation and a conjugate Fourier series method are used. For calculating the composite index, an effective-index method is described. After this treatment of the linear properties, the dispersion of the nonlinear properties is presented.

2. EFFECTIVE-INDEX APPROACH

The Lorentz-Lorentz equation relates the index of refraction to the polarizability of the molecule. This is the optical analog of the Clausius-Mossotti equation,⁶ which relates the low-frequency dielectric function to the polarizability:

$$\frac{n^2 - 1}{n^2 + 2} = \frac{4\pi}{3} N\alpha. \quad (1)$$

Here α is the microscopic polarizability, n is the index of refraction, and N is the number of molecules per unit volume and is equal to $A\rho/M$, that is, Avogadro's number times the density, divided by the molecular weight. Our goal is to determine the composite index of a polymer-chromophore system. Depending on the concentration of chromophores in the polymer, we assign the proper number of repeat units of the polymer, that is, an oligomer consisting of the set of repeat units and one molecule of the chromophore. Then, in a unit volume, the number of chromophores, the number of oligomers without chromophore, and the number of oligomers including the chromophore are all the same. Consequently the effective po-

Nonlinear Optical Properties Of Polymers

San-Hui Chi



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Nonlinear Optical Effects in Organic Polymers J. Messier, P. Prasad, D. Ulrich, 2012-12-06 Photonics the counterpart of electronics involves the usage of Photons instead of electrons to process information and perform various switching operations Photonics is projected to be the technology of the future because of the gain in speed processing and interconnectivity of network Nonlinear optical processes will play the key role in photonics Where they can be used for frequency conversion optical switching and modulation Organic molecules and polymers have emerged as a new class of highly promising nonlinear optical materials Which has captured the attention of scientists world wide The organic systems offer the advantage of large nonresonant nonlinearities derived from the π electrons contribution femtosecond response time and the flexibility to modify their molecular structures In addition organic polymers can easily be fabricated in various device structures compatible with the fiber optics communication system The area of nonlinear optics of organic molecules and polymers offers exciting opportunities for both fundamental research and technologic development It is truly an interdisciplinary area This proceeding is the outcome of the first NATO Advanced Research Workshop in this highly important area The objective of the workshop was to provide a forum for scientists of varying background from both universities and industries to come together and interface their expertise The scope of the workshop was multidisciplinary with active participations from Chemists physicists engineers and materials scientists from many countries Nonlinear Optical Properties of Polymers Alan J. Heeger, Joseph Orenstein, Donald R. Ulrich, 1988 **Nonlinear Optical and**

Electroactive Polymers Paras N. Prasad, D.R. Ulrich, 2012-12-06 This treatise is a compendium of papers based on invited talks presented at the American Chemical Society Symposium on Electroactive Polymers which covered nonlinear optical polymers and conducting polymers the common denominator being the correlated π electron structures The improved understanding of the consequences of π electron delocalization upon nonlinear optical properties and charge carrier dynamics has laid the foundation for the rapid development and application of the electroresponse of conjugated polymers As a result the area of electroactive and nonlinear optical polymers is emerging as a frontier of science and technology It is a multidisciplinary field that is bringing together scientists and engineers of varied background to interface their expertise The recent explosion of interest in this area stems from the prospect of utilizing nonlinear optical effects for optical switching and logic operations in optical computing optical signal processing optical sensing and optical fiber communications Polymers and organics are rapidly becoming one of the major material classes for nonlinear optical applications along with multiple quantum wells ferroelectrics and other oxides and direct band gap semiconductors The reasons for this lie in the unique molecular structures of polymers and organics and the ability to molecularly engineer the architecture of these structures

through chemical synthesis **Nonlinear Optics of Organic Molecules and Polymers** Hari Singh Nalwa, Seizo Miyata, 2020-07-09 The field of nonlinear optics emerged three decades ago with the development of the first operating laser and the demonstration of frequency doubling phenomena These milestone discoveries not only generated much interest in laser science but also set the stage for future work on nonlinear optics This book presents an excellent overview of the exciting new advances in nonlinear optical NLO materials and their applications in emerging photonics technologies It is the first reference source available to cover every NLO material published through 1995 All theoretical approaches measurement techniques materials technologies and applications are covered With more than 1 800 bibliographic citations 324 figures 218 tables and 812 equations this book is an invaluable reference source for graduate and undergraduate students researchers scientists and engineers working in academia and industries in chemistry solid state physics materials science optical and polymer engineering and computational science Structural Characterization and Linear and Nonlinear Optical Properties of Polymer Materials A. K. M. Matior Rahman, 1991 **Polymers for Second-order Nonlinear Optics** Geoffrey A. Lindsay, American Chemical Society. Meeting, 1995 Presents the most recent developments in second order nonlinear optical polymers Covers the most important technologies necessary to achieve commercially viable devices based on special polymeric materials with second order nonlinear optical properties Discusses important molecular design considerations how to process the polymers into films the stability of the films their optical properties and prototype devices that can be made from these films *Nonlinear Optical Properties of Organic and Polymeric Materials* David J. Williams, 1983 Good No Highlights No Markup all pages are intact Slight Shelfwear may have the corners slightly dented may have slight color changes slightly damaged spine **Optical and Electrical Properties of Polymers: Volume 214** John A. Emerson, John M. Torkelson, 1991-10-18 The MRS Symposium Proceeding series is an internationally recognised reference suitable for researchers and practitioners *Effect of Order on Linear and Nonlinear Optical Properties of Conjugated Polymers* Khashayar Pakbaz, 1993 Nonlinear Optical Properties of Liquid Crystals and Polymer Dispersed Liquid Crystals Francesco Simoni, 1997 This monograph is devoted to a detailed treatment of the nonlinear optical properties of liquid crystals The basic concepts of director optical reorientation and thermal nonlinearities are presented showing the fundamental theoretical approaches and describing the main experimental observations The presentation is self consistent and tutorial although the subject matter is of current research interest The last part of the book deals with more recent results on new composite materials Polymer Dispersed Liquid Crystals PDLC A general presentation of the optical properties is given and the observations of several nonlinear optical effects are reported Third-order Nonlinear Optical Properties of Conjugated Polymers and Blends San-Hui Chi, 2009 **Nonlinear Optical Properties of Organic Materials VI**, 1993 *Nonlinear Optical Properties of Organic Materials VII*, 1994 *Nonlinear Optical Properties of Organic Materials V* David J. Williams, 1993 *Nonlinear Optical Properties of Organic Materials VIII*, 1995 **Theoretical Studies of Nonlinear**

Optical Properties of Conjugated Polymers, 1993 The study of the non linear optical properties of polymeric systems is a challenging and exciting field of research ranging from device engineering optical measurements chemical synthesis to design and theoretical issues At the present time most of the basic science needed for the synthesis of molecules and the design of devices utilizing second order optical susceptibilities is in hand although certain issues remain to be resolved On the other hand many important questions regarding the design and use of third order optical susceptibilities are still unanswered The earliest ideas of the importance of low dimensionality optical band gaps suggests the use of conjugated molecules At present there is considerable effort both experimentally and theoretically in optimizing the value of gamma for polymers or oligomers with conjugated segments because such conjugated polymers like polyacetylene polythiophenes and the poly diacetylenes have very large gamma These polymers have also been under intense scrutiny because of their large conductivities when doped Although we are beginning to understand the theoretical reasons for the various unusual properties of the materials we do not understand the factors that limit the ultimate value of gamma For example what are the important structures and interactions in the molecule the prevent gamma from being as large as possible while still having a small absorption coefficient and how can we design molecules with these constraints in mind *Polymers for Photonics Applications II* Kwang-Sup Lee, 2003-07-03 The future of information technology requires ultra high speed processing and large data storage capacity Since the electronics technology using semi conduc tors and inorganic materials is about to reach its limits much current research is focused on utilizing much faster photons than electrons namely photonics To achieve any significant effect on the actual use of the science of photonics devel opments of more efficient photonics materials better optical property evaluations manufacture of devices for system applications etc are the subjects which need to be explored In particular the development of photonics materials stands in the forefront of research as this constitutes the most pertinent factor with regard to the development of ultra high speed and large capacity information processing In this respect there has been continuous research on photo responsive materials through molecular structure design and architecture and the results so far are very promising as functions and performances are beginning to realize their high expectations The two special volumes *Polymers for Photonics Applications* give authorita tive and critical reviews on up to date activities in various fields of photonic poly mers including their promising applications Seven articles have been contributed by internationally recognized and they deal with polymers for second and third order nonlinear optics quadratic parametric interactions in polymer waveguides electroluminescent polymers as light sources photoreflective polymers for holo graphic information storage and highly efficient two photon absorbing organics and polymers *Nonlinear Optical Properties of Semiconducting Polymers*, 1990 This program focused on the nonlinear optical properties of semiconducting conjugated polymers Specific emphasis was on fast time picosecond regime measurements of the photoconductivity of semiconducting polymers and on NLO measurements related to the mechanism and origin of the nonlinearity on attempts to optimize the magnitude of the

nonlinear response and on developing novel methods for measurement In the NLO area we succeeded in demonstrating that for conjugated polymers the nonlinear response is highly anisotropic with substantial nonlinearity only when the pump is polarized along the direction parallel to the polymer backbone In attempting to sort out the NLO mechanism we carried out comparative studies on cis and transpolyacetylene The larger NLO response from the trans isomer larger by at least a factor of 20 shows the importance of the degerate ground state *Nonlinear Optical Properties of Materials* Rashid A.

Ganeev,2013-01-09 This book is mostly concerned on the experimental research of the nonlinear optical characteristics of various media low and high order harmonic generation in different materials and formation and nonlinear optical characterization of clusters We also demonstrate the inter connection between these areas of nonlinear optics Nonlinear optical properties of media such as optical limiting can be applied in various areas of science and technology To define suitable materials for these applications one has to carefully analyse the nonlinear optical characteristics of various media such as the nonlinear refractive indices coefficients of nonlinear absorption saturation absorption intensities etc Knowing the nonlinear optical parameters of materials is also important for describing the propagation effects self interaction of intense laser pulses and optimisation of various nonlinear optical processes Among those processes one can admit the importance of the studies of the frequency conversion of coherent laser sources The area of interest for nonlinear optical characterization of materials is also closely related with new field of nanostructures formation and application during laser matter interaction We show how the nonlinear optical analysis of materials leads to improvement of their high order nonlinear optical response during the interaction with strong laser fields Ablation induced nanoparticles formation is correlated with their applications as efficient sources of coherent short wavelength photons From other side recent achievements of harmonic generation in plasmas are closely related with the knowledge of the properties of materials in the laser plumes All of these studies are concerned with the low order nonlinear optical features of various materials The novelty of the approach developed in present book is related with inter connection of those studies with each other

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