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Nonlinear Optical Materials

With 77 Illustrations

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FOREWORD

This IMA Volume in Mathematics and its Applications

NONLINEAR OPTICAL MATERIALS

is based on the proceedings of a very successful one-week workshop with the same title. This workshop was an integral part of the 1995–1996 IMA program on "Mathematical Methods in Materials Science." We would like to thank Jerome V. Moloney and John Sipe for their excellent work as organizers of the meeting. We would like to express our further gratitude to Moloney for editing the proceedings.

We also take this opportunity to thank the National Science Foundation (NSF), the Army Research Office (ARO) and the Office of Naval Research (ONR), whose financial support made the workshop possible.

Avner Friedman

Robert Gulliver

PREFACE

The search for materials, suitable for nonlinear optics technology applications, poses a great variety of challenges from a fundamental theoretical perspective. The IMA "Workshop on Nonlinear Optical Materials," held from February 4 – 8, 1996, provided the applied mathematics community with a flavor for the diverse applications areas that can be served if suitably optimized nonlinear optical material can be developed. The overall theme of the Workshop was twofold:

(i) the challenges faced in computing and optimizing nonlinear optical material properties starting either from first principles theories or suitably reliable phenomenological models and, (ii) the exploitation of these properties in important applications areas.

The properties of materials that makes them suitable for nonlinear optics applications are not those facing the traditional materials scientist. Familiar properties such as elasticity, hardness, etc. of a material may play an indirect role in terms of establishing the lifetime of a suitable optical material. However, the essence of a good nonlinear optical material lies in the magnitude of the so-called "nonlinear optical response function" or "nonlinear dielectric susceptibility," which provides a measure of how the material reacts to modify an incoming light beam. The latter quantity can be partitioned somewhat arbitrarily into a resonant or nonresonant optical response. Resonant optical response can involve either light absorption or amplification due to coupling to the dipole oscillators making up the material. A nonresonant optical response involves essentially no modification to the amplitude of the impinging light signal but, rather, induces an intensity dependent phase change. Hence the refractive index of the material is modified. In reality both absorption/amplification and refractive index changes occur simultaneously but one or the other can be enhanced depending on the application in mind.

For most nonlinear optics applications, the nonlinear optical response of a material arises from the collection of dipoles induced by the external optical field. These induced dipoles act in concert as sources in the Maxwell equations which describe the modification of the propagating light field. Computation of the appropriate nonlinear optical response function offers a major challenge requiring the solution of a complicated quantum mechanical many-body problem. The articles by Von Axt and Mukamel and Knorr and Koch, deal with the technical issues involved in truncating an infinite hierarchy of many-body equations so as to yield a reliable nonlinear optical response function for an important class of conjugated polymer systems and semiconductors. The latter are vital materials due to their relatively large induced dipole moments which translates into useful second order ($\chi 2$) and third-order (χ^3) nonlinear dielectric susceptibilities for nonresonant interactions. Resonant and near-resonant optical interac-

tions are vital in the operation of lasers and passive optical switches. The canonical example of a resonant interaction is the simple two-level atom where an incident optical wave can induce quantum mechanical transitions between the two discrete quantum states. If the quantum system is initially prepared with all of its population in the upper level (inverted state), then the system will amplify any incident propagating near-resonant light field. This is the basis for building optical amplifiers and lasers. Semiconductor lasers and amplifiers have made a huge impact in modern technology due to their intrinsically large resonant optical response which makes them suitable for integration into extremely small devices. Quantum confinement as in Quantum Wells (2D), Quantum Wires (1D) and Quantum Dots (0D), further enhances the optical nonlinearity of the semiconductor material. Very few such many-body calculations are feasible however even with present-day state-of-the art supercomputing technology.

A more practical approach involves the use of phenomenological models derived as coarse approximations of the more fundamental first-principles theories, with the relevant coefficients deduced directly from experimental measurement. Crystal symmetry plays a vital role in determining the generic form for the optical response function for nonresonant interactions. For materials lacking a center of symmetry, for example, it can be easily shown that the leading nonlinear term arises as a quadratic term in the optical field. This gives rise to the χ^2 dielectric response important in optical three-wave mixing processes, such as second harmonic generation (SHG) and frequency up-and down-conversion. For a material with a center of symmetry, the leading order nonlinear term is cubic in the electric field (χ^3) and this four-wave interaction gives rise to the optical Kerr effect, third harmonic generation (THG) and a variety of four-wave mixing (FWM) interactions. In many instances, the intrinsic optical nonlinearities are so weak that some form of intervention is needed to considerably enhance the strength of the interaction with a light field.

The remarkable flexibility available to enhance the intrinsic optical nonlinearity is reflected in a number of articles in this volume. We already mentioned above that quantum confinement in semiconductors can enhance the nonlinear optical response by many orders of magnitude. Knorr and Koch discuss novel nonlinear effects associated with ultrashort optical pulse propagation through stacks of Quantum Wells. Other techniques involve the addition of dopants to a host material and/or the use of certain composites to achieve strong enhancement of the nonlinear optical response. The article by Bergman and Levy discusses the use of a mix of a nonlinear dielectric and a metallic or semiconducting component to greatly enhance the local field and build an optical bistable switch operating at low external optical fields. Another approach involves the introduction of a periodic modulation of the material's linear and/or nonlinear refractive index. The resonant enhancement introduced by the nonlinear Bragg grating and its effect on propagating light fields is discussed in the article by de Sterke.

Here the additional controllable dispersion introduced by the grating structure can enhance second-order (χ^2) and third-order (χ^3) interactions and additionally lead to the formation of localized non-propagating stationary optical pulses called grating or Bragg solitons. Composites having fractal dimensions can lead to a many-order-of-magnitude increase of the nonlinear optical dielectric response by causing a large enhancement of the local field as discussed by Stockman. A particularly novel approach to enhancement of the nonlinear dielectric response of a material is to apply a magnetic field to the material. Boardman and Xie discuss how the magnetooptic effect can be exploited to achieve polarization control in nonlinear optical waveguides and enhance the formation of χ^2 optical solitary waves.

Another important consideration when seeking suitable optical materials is the issue of the potentially huge separation in time and space scales. If one considers an incident light field as a probe of a material, one has the remarkable situation that this interaction may involve a continuous low intensity optical wave (CW) all the way down to a few femtosecond (10^{-15} second) optical pulse. This enormous range of accessible timescales means that very different physics may be operative during the interaction process. For example, very short optical pulses are likely to be particularly sensitive to the intrinsic material dispersion. Linear dispersion of the glass in an optical fiber core is responsible for the formation of optical fiber solitons in long distance communications fibers. Here the optical pulses are relatively long ($\approx$ 20 picoseconds) and the weak positive Kerr optical nonlinearity balances the weak anomalous dispersion of the material. Ultrashort femtosecond duration optical pulses have very large initial frequency bandwidths and the added nonlinear self-phase modulation accumulated during propagation causes additional huge spectral superbroadening. Add to this the likelihood of optical shock formation at the high peak intensities achievable in such ultrashort optical pulses and it becomes clear that the material's nonlinear dispersion will play an important role in regularizing the system behavior. The article by Sheik-Bahae discusses recent developments in determining the nonlinear absorption and dispersion of a wide range of optical materials. The essential result here is a generalization of the Kramers- Kronig (Hilbert transform) relation to deal with the third-order (χ^3) nonlinear optical response. Even though ultrashort optical pulses may have very high local intensities, the pulse itself may have very low energy (time and space integrated pulse intensity). High intensities in bulk materials can lead to material breakdown with possible catastrophic consequences. In fact laser-induced breakdown is one of the main limitations in the development of efficient nonlinear optical devices. While the threshold for LIB depends on the local intensity of the light field, the latter intensity threshold can be reached as a consequence of critical self-focusing which leads to an explosive growth in the light intensity at the nonlinear self-focus. Mathematically, this is simply the self-similar critical collapse associated with the higher dimensional nonlinear Schrödinger (NLS) equa-

tion. Associated with critical collapse is the dramatic narrowing of the light beam as the collapse singularity is approached. This must mean that the NLS envelope description must lose its validity and a full vector Maxwell description is required. The article by Moloney offers an overview of very recent developments in the study of ultrashort pulse propagation in bulk optically transparent materials. While initially transparent, the formation of a plasma above a critical intensity introduces a local dissipation which halts the critical collapse.

There is little doubt that nonlinear optics, whether realized in an all-optical or a hybrid electro-optical mode, will have an increasingly important impact on modern technology. The search for suitable materials to satisfy future technology needs is an ongoing challenge. Can mathematics contribute usefully to this important emerging area? Clearly there are issues with regard to establishing useful scaling laws for estimating the size of the optical nonlinearity. The origin of the optical nonlinearity depends on the relevant time scale of the light interaction with the material. For ultrafast processes in the femtosecond time domain, distortion of the atom's or molecule's electron cloud leads to an induced electronic nonlinearity which is weak but very fast. Longer time interactions, for example in the picosecond domain, can lead to coupling with the intrinsic vibrational nuclear motions in a molecule or can reorient small molecules, leading to a stronger but slower, nonlinear coupling. The vibrational resonant processes, although highly detuned from the very large optical frequency of $\approx 10^{15}$ second^1 (their resonances lie around 10^{13} second^{-1}), can be driven through a three-wave interaction involving the original incident optical wave, the above material oscillation and a spontaneously generated scattered optical wave (Stimulated Raman Scattering (SRS)). Interaction times of the order of nanoseconds (10^{-9} seconds) can stimulate hypersonic acoustic waves via a similar three-wave interaction called Stimulated Brillouin Scattering (SBS). Even longer interaction times can induce thermal coupling which poses a major problem in CW optical devices such as semiconductor lasers, amplifiers and repetitively pumped pulse systems.

Jerome V. Moloney

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