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Nonlinear-Optical studies of organic liquids and polymer optical fibers

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Abstract

The field of experimental nonlinear optics was born with the observation of second harmonic generation by Franken *et al.* wherein coherent, high-intensity laser light having a center wavelength of 694 nm, impinging on a sample of crystalline quartz produced radiation at twice the frequency (or half the wavelength, i. e., 347 nm) of the incident radiation. Shortly thereafter, observation of a remarkable two-photon process in which radiation incident on an isotropic sample that was seemingly transparent to it produced a fluorescence having a wavelength much shorter than the incident radiation.

Keywords: Nonlinear-Optical studies, organic liquids, polymer optical fibers.

Introduction

Fluorescence was attributed to two-photon absorption, wherein the simultaneous absorption of two photons by the medium resulted in an excitation to an excited state whose fluorescence decay was observed. The fluorescence was quadratically dependent on the incident intensity.

While early theoretical work concerning nonlinear-optical processes, like two-photon absorption, had been done, it was the advent of the laser (or optical maser as it was originally called), which led to the aforementioned experimental observations. These and other studies have stimulated further development of nonlinear optical theory and experiment that has continued unabated to date. It is no accident that the birth of experimental nonlinear optics closely followed the experimental realization of the laser.

Excitation of nonlinear-optical responses in materials generally requires radiation that has both a high field strength and a high degree of phase coherence. In many instances, lasers can be so efficient at producing nonlinear effects that they can lead to catastrophic damage of either the lasing medium or other components of the laser or experimental system.

It is interesting to note that while much current research emphasizes materials with a maximized nonlinear-optical response, many early investigations of the nonlinear-optical properties of lasing media were concerned with the mechanisms leading to self-destructive effects, which are minimized in materials with small nonlinearities.

The nonlinear-optical (NLO) response of materials is a means for the ends of both pure and applied research. From the point of view of basic research, the NLO response of a given system can be used to provide experimental conditions not attainable by other means. A simple example of this is harmonic conversion of laser radiation from standard, well-developed laser sources, like NdYAG, to frequencies where laser radiation is not directly accessible. (For example, this is the method to be applied on a large scale at the National Ignition Facility being built to study inertial confinement fusion. Light from (large) NdYAG lasers (1064 nm) will be frequency tripled to 354.7 nm before being focused onto the target.) Of course, beyond their experimental usefulness, the NLO properties and responses of a medium can shed light on the nature of the medium itself. For example, different types of NLO spectroscopy can be used to probe energy eigen states, transitions, and other bulk and molecular properties of a medium that are not accessible by other spectroscopic methods. A close wedding of theoretical modeling and experimental measurement can result in a better understanding of the fundamental nature of the system as manifested in its NLO response.

The non-interaction of photons is in stark contrast to the unceasing interaction of the

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electrons used to make electronic switches. Their interaction is a double-edged sword; it makes them relatively easy to control, but it also makes the response of devices based on their control relatively slow. This limitation becomes ever more apparent as the sizes of electronic control devices (switches, CPUs, etc.) become ever smaller. What is needed is a device in which the information-carrying elements move unimpeded from place to place (with a minimum of interaction) and can be switched very quickly when they need to be re-routed.

Modern telecommunications networks are composed of two main elements, long-haul optical fibers that pipe information very quickly over long distances, and electrical amplifiers and switches. It is the conversion from light to electrical current that is the main speed bottleneck in the system. The all-optical switch is the crux of the all-optical network.

Depending on the mechanism responsible for the NLO response of the system, all-optical switches can be very fast (fem to second time scale) and thus can switch signals much faster than electrical switches (nanosecond time scale). While it can be argued that all-optical switching has been achieved in the laboratory, the current state of the art in NLO research is not capable of producing a commercially viable ultrafast all-optical switch. The most straightforward solution to producing an all-optical switch is to construct it out of a material that has a large nonlinearity. Materials with large nonlinearities require less power to excite the NLO processes that are necessary for switching action in the material.

Research Study

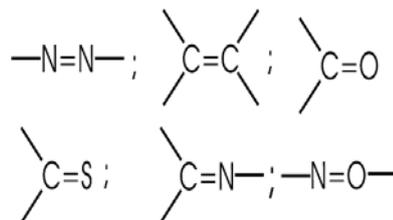
The nonlinear optical properties of organic systems are of interest because, unlike inorganic systems, where the bulk nonlinear optical phenomena arise from band structure effects, the bulk nonlinear optical response of organic systems originates in the virtual electron excitations occurring on the individual molecular scale. The origin of the potentially large nonlinear-optical response of certain organic systems is the relative ease with which optical fields affect the motions of the π -electrons that take part in the delocalized covalent bonds characteristic of unsaturated organic compounds.

Because the electrons in these so-called π -conjugated systems are not tightly bound to the individual nuclear sites, their orbital motion can extend over long distances, spanning the entire molecule or even macroscopic length scales in bulk samples.

Computational quantum chemistry (CQC) methods (both ab initio and semi-empirical) are increasingly used to augment more analytical theoretical methods to guide the researcher in predicting the polarizabilities associated with a particular molecule.

CQC methods can also shed light on the physical underpinnings of the molecular NLO properties. For example, CQC calculations have shown the need to account for electron correlation effects (i.e., the fact that electrons in a molecular system react to one another's motion and attempt to keep out of each other's way) when calculating the NLO properties of molecules.

CQC calculations, analytical calculations and experimental results have also shown that, in general, two-photon states are necessary to fully describe the third-order molecular NLO response. Similar agreement has been obtained



between theory and experiment for the sign of the third-order response for certain molecular systems. This is particularly important, because the sign of the nonlinearity is an important factor in determining the consequences of the nonlinear refractive index—a positive nonlinear refractive index can lead to catastrophic self-focusing in a medium, whereas a negative nonlinearity leads to the opposite effect of self-defocusing. Because the large number atoms in typical molecules (even small ones), computational quantum chemistry is still at an intermediate stage of development from a quantitative standpoint; but it is already an indispensable tool. It will only become more valuable as computational speed, power, and theory progress.

It has long been known that conjugated molecules (i.e., molecules with alternating single and double bonds) possessing certain groups, called chromophores, have very strong absorption bands in the visible and ultraviolet regions of the spectrum.

Figure shows some examples of chromophores. This conjugation strongly affects the molecular polarizability α , which determines the molecular absorption spectrum. It does not seem unreasonable to postulate if the conjugation affects the linear optical properties of a molecule it might also have an appreciable effect on its nonlinear-optical properties. There is a vast literature on NLO studies of organic molecules, much of it concerned with the structure property relationships necessary to optimize the molecular NLO response. Organic dyes are a subset of conjugated organic molecules, and are promising optical materials due to their relatively large dipole transition moments and strong, narrow absorption features.

While their strong absorption precludes the use of devices made using these dyes in their one-photon resonant regime, we will show that the study of their NLO spectra in the resonant regime can give information that is useful in determining higher-lying excited states, the knowledge of which are necessary to accurately predict the molecular NLO response at other wavelengths.

Theory of the Microscopic Nonlinear Susceptibility

The response of ponderable media to an electromagnetic field \mathbf{F} (\mathbf{E} , \mathbf{B}) is described by the temporal and spatial evolution of the displacement and magnetic fields \mathbf{D} and \mathbf{H} induced in the medium by the vacuum electric and magnetic-flux density fields \mathbf{E} and \mathbf{B} . These induced fields are due to the motion of the free and bound charges that comprise the medium. The constitutive relation between the vacuum fields \mathbf{E} and \mathbf{B} and the 'medium' fields \mathbf{D} and \mathbf{H} give a complete description of the system response in terms of the vacuum fields.

We are particularly interested in the relationship between the displacement field and the electric field

$$\mathbf{D} = \mathbf{E} + 4\pi\mathbf{P},$$

where the electronic polarization \mathbf{P} describes the radiation field due to the response of the bound charges in the medium to the vacuum electric field.

Optical Kerr Effect Data Analysis

Four types of optical Kerr effect measurements were performed:

- Referenced measurements—where the third-order response of an unknown liquid was measured with respect to that of a standard, previously characterized liquid
- Absolute measurements—all experimental parameters necessary were measured to calculate the absolute value of the third-order susceptibility from equation.
- Time-delay studies—where the time-dependence of the sample's third-order response was measured.
- Dispersion measurements that determined the frequency response of the third-order susceptibility of the sample.

Referenced Measurements

Measurements were performed that measured the absolute value of the optical Kerr susceptibility of a previously uncharacterized neat liquid or solution with respect to that of a well-characterized liquid like nitrobenzene (C₆H₅NO₂) or carbondisulfide (CS₂) in real-time. The meaning of 'real time' is that the reference material was placed in one arm of the apparatus and the 'unknown' sample was placed in the other arm. This type of referencing automatically corrects for different signals arising from shot-to-shot fluctuations in the spatial and temporal profiles of the pump and probe pulses and other in-common parameters. Because the optical Kerr signal is fairly sensitive to small changes in the parameters on which it depends, referencing can substantially reduce measurement error, but if the fluctuations are too severe, referencing will be of little help.

Photomultipliers

The responsivity of the PMTs was both calculated from the Burle specification sheet and measured. The photocathode spectral response is very flat in the region 400 nm–800 nm. Table lists the relevant parameters for two different tube voltages. The tube responsivity is calculated by multiplying the photocathode responsivity by the current gain at the operating voltage. The PMT responsivity was also measured by exposing the PMT to a very weak light-emitting diode source, and viewing the PMT's current pulse on a digital oscilloscope with a 50 Ω input impedance. The circuit that powered the LED was constructed to produce a very low current. Since the current through the LED circuit is approximately proportional to the number of photons emitted by the LED per unit time, if the current is small enough, only a single photon will hit the PMT photocathode during the time corresponding to a single oscilloscope sweep (50 ns).

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