Aug 9th, 10:15 AM - 12:00 PM

Second hyperpolarizability of carbon tetrachloride

Phillip C. Lotshaw  
Willamette University

Anna M. Smith  
Davidson College

David P. Shelton  
University of Nevada, Las Vegas, shelton@physics.unlv.edu

Repository Citation  
http://digitalscholarship.unlv.edu/cs_urop/2011/aug9/31

This Event is brought to you for free and open access by the Undergraduate Research at Digital Scholarship@UNLV. It has been accepted for inclusion in Undergraduate Research Opportunities Program (UROP) by an authorized administrator of Digital Scholarship@UNLV. For more information, please contact digitalscholarship@unlv.edu.
Second Hyperpolarizability of Carbon Tetrachloride

Phillip C. Lotshaw¹, Anna M. Smith², and Dr. David P. Shelton³

¹Department of Physics, Willamette University, Salem, OR 97301, USA
²Department of Physics, Davidson College, Davidson, NC 28035, USA
³Department of Physics, University of Nevada-Las Vegas, Las Vegas, NV 89114, USA, NSF-REU Program Summer 2011

Background

Although present theories of nonlinear optics agree with observed behavior in simple atoms such as helium, more complex molecules containing many electrons, such as carbon tetrachloride (CCI₄), cannot consistently be described by theory. Through experimental analysis of nonlinear materials, a new, more sophisticated model for describing their properties could be realized. The purpose of our experiment was to

In the presence of a static electric field E₀ and an electric field E₀ varying with frequency ω₀, the term is given by:

\[
\frac{\Delta E}{\Delta t} = \frac{1}{\epsilon_0} \left[ \nabla \times \vec{E} \right] \cdot \vec{H} - \frac{1}{\mu_0} \nabla \times \vec{H} \times \nabla \Phi
\]

Materials with nonlinear properties can convert light from one frequency to another, allowing for generation of many different frequencies from a single source. We determined the second-harmonic generation, a nonlinear process in which two light waves of equal frequency are combined into a single light wave with double the frequency of either initial wave. The experiment utilized static electric field induced single-mode (ESHG) to produce a measureable second-harmonic signal from CCl₄. The signal was compared with the signal from nitrogen, a gas of known second hyperpolarizability, to determine the value of γ for CCl₄. Our measured value was compared to previously measured values at varying frequencies and with the prediction of the CCSD(T) mathematical model using the 31G(3d)+pd basis set, the highest approximation by reference 2.

The relevant equation for the second hyperpolarizability is given by the Taylor series expansion of the dipole moment with respect to the local electric field E:

\[
\mu_{\text{elas}} = \mu_0 + \alpha E^2 + \beta E^4 + \gamma E^6 + \ldots
\]

In the presence of a static electric field E₀ and an electric field E₀ varying with frequency ω₀, the term is given by:

\[
\frac{\Delta E}{\Delta t} = \frac{1}{\epsilon_0} \left[ \nabla \times \vec{E} \right] \cdot \vec{H} - \frac{1}{\mu_0} \nabla \times \vec{H} \times \nabla \Phi
\]

Figure 1: The static electric field generated by the electrode array is determined by the spacing between the electrodes. The array used in our experiment consisted of 150 pairs of electrodes separated by 2.692 mm.

Typically, the second-harmonic wave and the fundamental wave travel at different speeds through the nonlinear medium. As the length of the medium is increased, the second-harmonic waves generated at the beginning and the end of the sample get progressively farther out of phase, interfering perfectly destructively at the coherence length of the sample, causing a phase shift of π radians in the generated second-harmonic wave, and therefore causing all second harmonic waves to be generated in phase with each other. The coherence length of the sample is adjusted to match the spacing of the electrodes by changing the density of the sample.

Figure 3: A schematic diagram of our experimental setup. Fused silica prisms allow for transmission of UV light.

Figure 4: The static electric field generated by the electrode array is determined by the spacing between the electrodes. The array used in our experiment consisted of 150 pairs of electrodes separated by 2.692 mm.

We performed our experiments by taking second-harmonic signal measurements for triplets of N₂ gas, a mixture of N₂ and CCl₄ gases, and N₂ gas. The decision to measure the nonlinear behavior of the second harmonic signal generated from CCl₄ and to compare the results with the prediction by the CCSD(T) molecular model.

Figure 5: Typical triplet data for absorption filters of N₂, N₂ and CCl₄, and N₂.

The peak signal value and pressure were approximated by a quadratic equation. Because each peak was not perfectly parabolic, a better approximation was obtained by only analyzing the top fifty percent of the data. This method also accounted for short power fluctuations of the laser by averaging over a range of pressures.

To further the study of the nonlinear properties of CCl₄ and other molecules, one would like to eliminate error due to power and mode fluctuations by testing with a more stable laser. Also, performing the experiment at multiple fundamental wavelengths will give a more accurate best fit line for "γ" versus ω₀. A more accurate γ value would provide a better foundation for refining current molecular models.

Acknowledgements

We would like to thank Dr. David P. Shelton for his extensive help with all aspects of the experiment. Support from the REU program of the National Science Foundation under grant DMR-1005347 is gratefully acknowledged.

References