A measurement of the second hyperpolarizability of carbon tetrachloride by ESHG at 514.5 nm

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Abstract

The second hyperpolarizability (γ) of carbon tetrachloride was measured by gas-phase electric-field-induced second-harmonic generation (ESHG). Periodic phase matching was used in order to enhance the ESHG by gases illuminated by a cw argon-ion laser operating at 514.5 nm. Using nitrogen as a reference gas, γ_{CCl4} can be calculated by means of the ratio $\gamma_{CCl4}/\gamma_{N2}$ determined to be 18.3 ± 0.7.

Setup

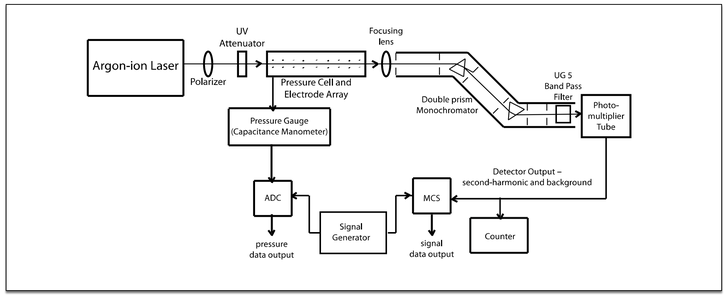

Fig. 1: Schematic diagram of the experimental apparatus, showing the cell in which the ESHG beam is produced, the monochromator which separates the second-harmonic beam, and the configuration of electronics which allows for the synchronous collection of pressure and signal data.

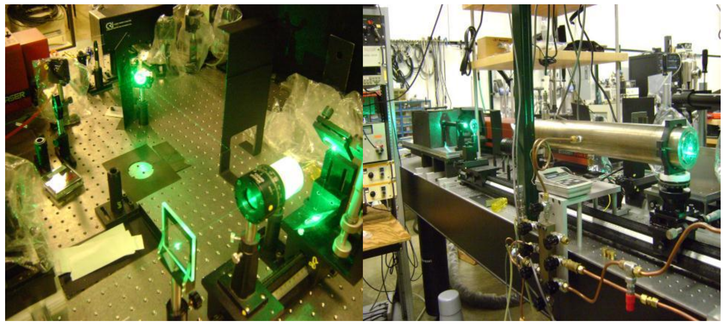

Fig. 2: Left: Upstream path of fundamental beam showing the polarizer, UV attenuator and various other optics. Right: Looking downstream: gas sample cell with the manifold and various valves used to control the pressure and composition of the cell.

Experimental Method

A schematic of the apparatus is shown in Figure 1. The fundamental beam is generated by an argon-ion laser. The beam then passes through the gas sample and electrode array inside of a stainless-steel pressure cell. An attenuator absorbs any coherent second-harmonic background that may have been produced by the optics leading up to the cell, see Figure 2. The electrode array provides a periodic static field which enables the production of the second-harmonic by the isotropic gas sample in response to the optical field.

One can maximize the ESHG signal by adjusting the pressure inside the cell in order to achieve phase matching such that the coherence length of the gas sample matches the electrode array spacing. The second-harmonic beam exits the cell colinearly with the fundamental beam and is separated from it by a double prism monochromator and a UG 5 band pass filter.

The signal is detected by a photomultiplier tube and individual photons are counted. A logic pulse generator is used as an external clock in order to simultaneously collect and record the second-harmonic signal and the voltages from a capacitance manometer as we scan the pressure inside the sample cell through phase match.

Analysis

The peak second-harmonic signal, $S^{(2\omega)}$, and the phase match density, ρ, it occurs at are determined by fitting raw data on SigmaPlot, see Figure 3.

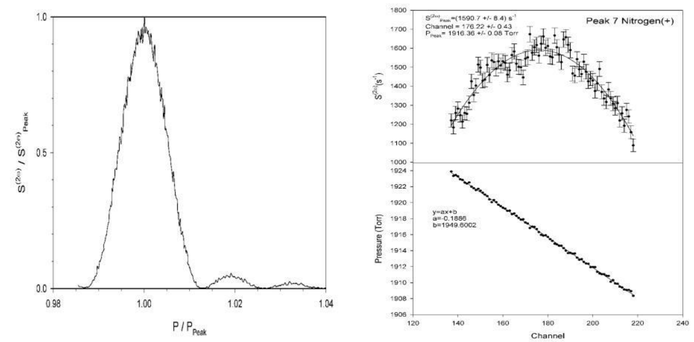

Fig. 3: Left: This plot shows the variation of the second-harmonic signal as the pressure in the cell was scanned through phase match. Right: An example of the fits used to determine peak signal and peak pressure.

Analysis (cont.)

By keeping experimental parameters constant between pure nitrogen and carbon tetrachloride-nitrogen mixture gas samples and making systematic measurements as to account for temporal drifts in signal, the ratio $\gamma_{CCl4}/\gamma_{N2}$ can be evaluated using the measured values of $S^{(2\omega)}$ and ρ from the relationship

$$\gamma \propto \frac{\sqrt{S^{(2\omega)}}}{\rho}.$$

Results

λ (nm)	ρ_{N2}/ρ_{CCl4}	$\gamma_{CCl4}/\gamma_{N2}$	γ_{CCl4}[a] (10^{-61} C^4 m^4 J^{-3})
514.5	11.4 ± 0.1	18.3 ± 0.7	13.4 ± 0.5

[a] Obtained using Ref. 2. γ_{N2} = 73.4 ±0.3 10^{-63} C^4 m^4 J^{-3}

Acknowledgements

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