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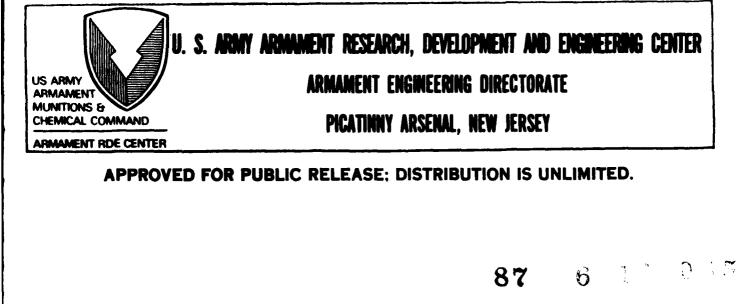
TECHNICAL REPORT ARAED-TR-87015

RELATIVE VALUE OF THE THIRD-ORDER NONRESONANT SUSCEPTIBILITY OF WATER



SUNG LA L. E. HARRIS





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INTRODUCTION

In the theory of Coherent Antistokes Raman Scattering (CARS) (refs 1 and 2), the signal is proportional to the squared modulus of the total third-order susceptibility $\chi^{(3)}$ which is the sum of the resonant susceptibility χ_r and nonresonant susceptibility χ_{nr} :

$$|x^{(3)}|^2 = (x^* + x_{nr})^2 + x^{2},$$

where χ' and χ'' are real and imaginary parts of the resonant susceptibility. It may be stated that the resonant susceptibility is related to the nuclear displacement and nonresonant susceptibility to electronic polarization. The cross term $\chi' \chi_{nr}$ which mixes resonant and nonresonant susceptibility modulates the line shape of the resonant spectrum. The effect becomes more pronounced as the number denisty of the resonant medium is lowered to a very low level relative to the nonresonant background. Theoretical compensation for this effect can be made from the known ratio of χ_{nr}/χ_{r} . Using (CARS) four wave mixing, the nonresonant third-order susceptibility of several gases was first measured by Rado (ref 3). His measurements are based on the reference value of hydrogen resonant susceptibility obtained from electric field induced absorption measurements. Employing essentially the same technique, Lundeen et. al. (ref 4) greatly extended the list of gases to include various hydrocarbons and halocarbons. Their reference value of the resonant susceptibility of H₂ was calculated from Raman cross section and line width data.

Third-order nonresonant susceptibility can be measured by other nonlinear processes; namely, electric field induced second harmonic generation (refs 5 and 6), dc Kerr effect (ref 7), and third-harmonic generation (ref 8). Recently. using a phase-modulation technique for coherent Raman spectroscopy, Rosasco and Hurst (ref 9) have determined the absolute value of the nonresonant susceptibility of Ar and renormalized the results of Rado by recalculating the value of the resonant H₂ susceptibility. As for the nonresonant susceptibility of water, at the present time there is only one reported value by Ward and Miller (ref 5). In particular, they have investigated the temperature dependence of field induced second-harmonic generation in the gas phase for molecules having a permament electric dipole moment. Effective nonresonant susceptibility is then obtained by extrapolating the inverse of temperature to zero. Their value for the ratio of nonresonant susceptibility of water to that of N_2 is 2.24. Levine and Bethea (ref 10) measured a similar quantity for liquid water and offered a means to extract the isolated molecular value from their data. The temperature dependence of the CARS resonant spectrum of water vapor and mixtures with other gases was investigated by Hall and Shirley (ref 11) and also by Farrow et. al. (ref 12).

EXPERIMENTAL

The apparatus used to generate the CARS signal is shown in figure 1. One of the main elements of the experiment is the pulsed, frequency doubled Nd:YAG laser (Quanta Ray DCR-1A). This beam is split into about two equal parts. One part is used as the pump beam ω_1 and the other is used to drive a Quanta-Ray PDL-1 dye laser that is operated broadband using Exiton dyes (LDS698 225 mg/1, DCM 5.8×10^{-5} M for the oscillator; LDS 698 115mg/1, DCM 0.6×10^{-5} M for the ampilifier in ethanol) to generate the Stokes beam ω_2 centered at 695 nm. The pump beam was attenuated to avoid optical breakdown. The energy was about 19 mj/pulse. The Stokes beam ω_p was combined collinearly with ω_1 using a long-pass dichroic mirror DM2. The combined beam was then focused into the temperature controlled sample cell with a lens Ll (140mm focal length) and recollimated by an identical lens L2. To optimize phase matching, a 12.5-mm thick optical flat rotatable about its vertical axis is inserted in the Stokes beam before focusing. The anti-Stokes beam ω_3 is separated from the pump and Stokes beam by a Shott color filter (BG25) and a prism. The signal is then diverted by a mirror to a condensing lens L3 which focuses the signal onto the entrance slit (100 um) of a 0.25-m monochromator equipped with a 1800 lines/mm grating. The signal is detected by a vidicon detector (PAR SIT) and processed by a PAR OMA 3 system.

The sample cell used for the experiment was obtained from Perkin Elmer (model IRI04). This model is a heated gas cell having 50-mm path length and 50mm diameter quartz windows. It can be heated to a set temperature up to 200°C at 30 psig. The cell has three ports. One is for connecting to a vacuum system and the second port is equipped with a rubber septum (10 mm diameter and 2 mm thick) through which a measured amount of liquid sample can be injected by use of a The third port was originally provided as a safety valve by microsyringe. installing an aluminum foil of predetermined rupture strength together with an inner layer of teflon. In this experiment, the third port was used for the purpose of injecting gas by installing an open-close valve. To remove any possible contaminants before using, the cell was simultaneously heated to 180°C and evacuated. When the pressure in the cell reached below 10^{-3} torr, the cell was disconnected from the vacuum system and cooled below 373 K (100°C). Distilleddeionized water was drawn into the syringe up to a predetermined level and emptied onto an electronic balance (Sartorius 1602 MP). The mass of water in the syringe can be reproduced within 3% error. Because of the high vacuum inside the cell, when the needle is injected into the septum, the water in the syringe is completely drawn into the cell. Therefore, the amount of water injected into the cell was considered the same as the measurement on the balance.

RESULTS

As a reference value for the third-order nonresonant susceptibility to be used with respect to that of water vapor, the nitrogen CARS signal was measured. The N₂ spectrum (fig. 2) represents the accumulated counts of 250 laser shots. The maximum intensity occurs at 4307 cm⁻¹. The full width at the half maximum is about 235 cm⁻¹. The calibration was done with Argon lines. The amount of N₂ in the cell can be readily calculated using the ideal gas law with known pressure, temperature, and cell volume. In this sample, the nitrogen content was 8.9×10^{-3} mole. CARS measurements for water were carried out as a function of temperature. The amount of water used in the measurement was chosen so that at 373 K (100°C) the vapor pressure does not exceed the saturation pressure at l atm, yet it provides sufficient number density for the measurement. The measured temperature dependence of CARS intensity at the peak is shown in The amount of water in the sample was 2.44×10^{-3} mole. A CARS signal figure 3. reaches maximum count near 413 K (140°C). Once again, this represents accumulated counts of 250 laser shots. The steady rise of CARS signal in the temperature range 383 to 423 K (100 to 150°C) is likely due to the condensation of water on the inner surface of the rubber septum whose temperature may lag appreciably behind that of the metal surface of the cell. Central regions of the two rather large windows of the cell may also have considerably lower temperature The possibility that there may exist a certain amount of associated as well. water vapor may safely be discounted. The calculated value of the ratio of dimer to monomer at 400 K (127°C) and at 1 atm is only 0.003 (ref 13). The water spectrum at 413 K (140°C) is shown on figure 4. The nonresonant spectrum is the same as that of nitrogen. The resonant peak at 3655 cm^{-1} coincides with the vibrational spectrum of water due to symmetric stretching reported at 3657 cm^{-1} (refs 11 and 12). Because the two peaks are well separated and the resonant peak has relatively weak intensity and small width, the effect of intermodulation may The effect is, therefore, not taken into be expected to be small. The ratio of nonresonant susceptibility of water to that of consideration. nitrogen was computed on the assumption that the CARS signal is proportional to the square of the number density. The value obtained is 2.27. The probable error in the measurement is estimated to be 12% primarily due to fluctuations in pump laser intensity. The corresponding value calculated from the individual values of water and nitrogen given by Ward and Miller (ref 5) is 2.24. The reported uncertainties in their individual values range from 2 to 5%. Therefore, the uncertainty in the ratio may be expected to be of the same order as the estimated error in these measurements.

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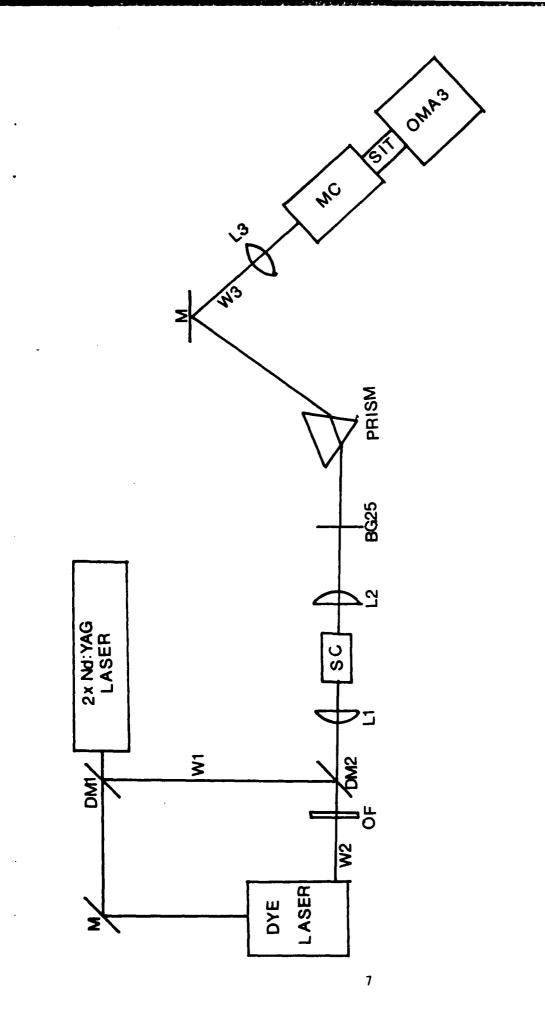
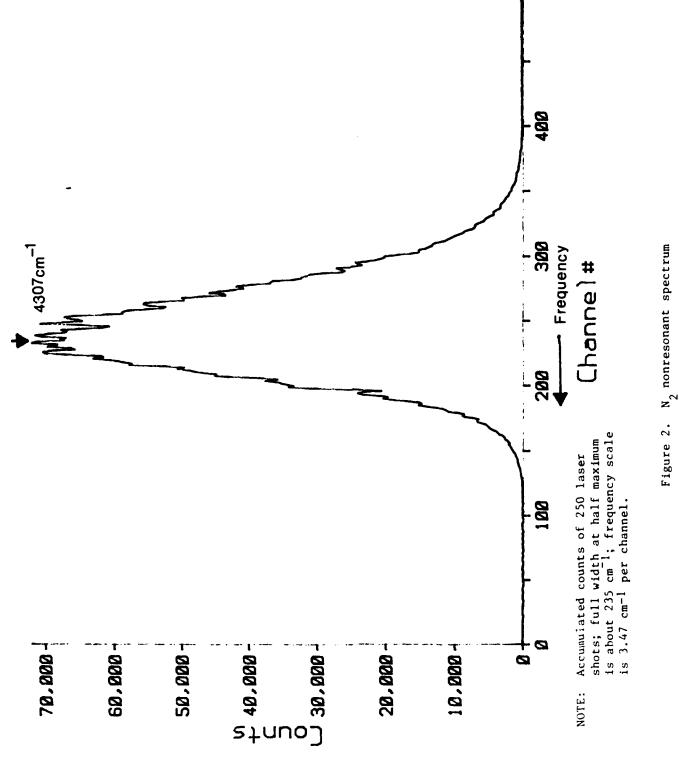


Figure 1. Schematic diagram of colinear CARS experiment

M = mirror OF = optical flat MC = monochromator SC = sample cell



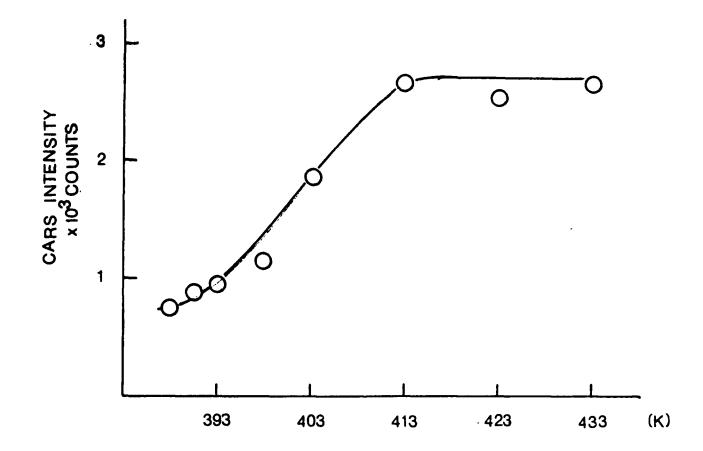
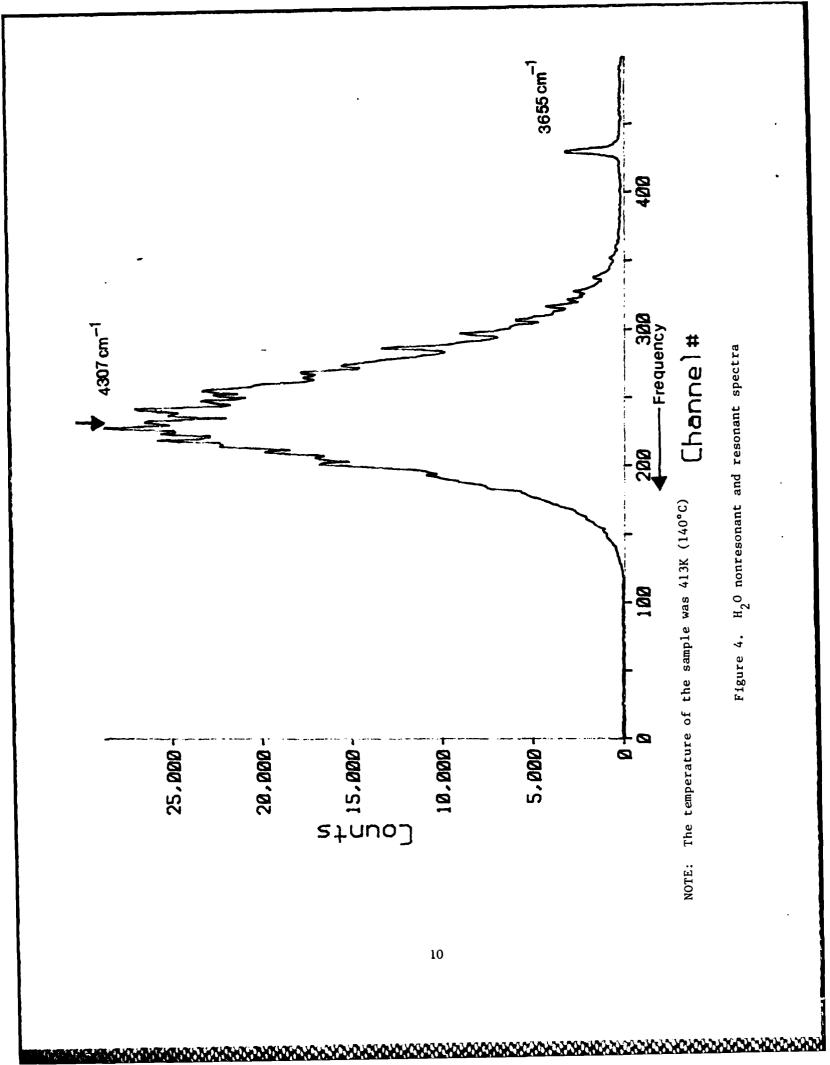


Figure 3. Temperature dependence of the CARS signal of water at the peak of the nonresonant spectrum



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