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A Continued Study of Optical Sound  
Generation and Amplification

Henry E. Bass and F. Douglas Shields

Physical Acoustics Research Laboratory  
University of Mississippi  
University, Mississippi 38677  
PARGUM-88-02

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19) Abstract (Continued)

Measurements ~~this year have been~~ <sup>are</sup> concerned with non linear dependence of the deflection of the detecting beam upon energy deposited in the liquid. The results ~~to date~~ indicate a change of state in the very small irradiated volume.

In the part of the project concerned with the propagation of sound through excited gases, an effort has been made to observe SACER in CO. Difficulty has been encountered in heating the vibrational mode in this gas with an electrical discharge. However, CO has been added to N<sub>2</sub> in small amounts to act as an indicator of the vibrational temperature in N<sub>2</sub>. Due to a relatively fast v-v transfer the vibrational modes of the two gases stay in equilibrium. The I.R. radiation coming from the I.R. active CO is then an indicator of the vibrational temperature of the mixture. ~~In this way~~ the decay of the vibrational temperature previously only inferred from the rise and fall of the translational temperature has been measured directly. This gives added confidence to conduction and relaxation times previously deduced from the translational-temperature vs time curves. An effort will now be made to measure vibrational relaxation times in H<sub>2</sub> and H<sub>2</sub> mixtures, which previously have not been measured.

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## Table of Contents

Report Documentation Page.....	I
1.0 Optoacoustic Studies of Liquids.....	1
1.1 Introduction.....	1
1.2 Experimental Configuration.....	3
1.3 Microscopic Interpretation of Optoacoustics in Fluids.....	5
References .....	20
2.0 Ultrasonic Attenuation in Polymer/Water Mixtures.....	22
3.0 The Propagation of Sound in Vibrationally Excited Gases (SACER)....	23
3.1 Summary of Past Work.....	23
3.2 Work during the Past Year.....	24
3.2a Bibliography of Previous Work with CO.....	25
3.2b Efforts at vibrationally exciting CO in an electrical discharge.....	28
3.2c Measurements of the vibrational temperature of N <sub>2</sub> using CO as a monitoring gas.....	28
3.3 Work Planned for the Coming Year.....	30
References.....	34
Appendix A: Publications/Patents/Presentations/Honors Report.....	35
Appendix B: Status Report.....	44

## 1.0 Optoacoustic Studies of Liquids

### 1.1 Introduction

Investigations of optoacoustic generation in liquids are well documented.<sup>1,2,3</sup> Most of these studies were carried out using low optical energies and small optical absorption coefficients. A small optical absorption coefficient makes the symmetry of the problem simple; the mathematics tractable. In the low optical energy regime, the acoustic generation mechanism is thermal expansion. This is the basis for linear optoacoustic theory. Nonlinear effects occur when the liquid experiences multiphoton absorption or a phase transition.

On a molecular level, the optoacoustic effect can be broken down into several different steps. The process of converting optical energy to acoustical energy begins with the absorption of photons by the molecules of the fluid. This absorption leaves the molecules in an excited state. In the present case, a nitrogen laser, used as the energy source, produces electronic excitation of the molecules. Dissipation of this energy can occur through several mechanisms including; photochemical processes, radiative decay, and nonradiative decay. In nonradiative decay, the electronic excitation energy is transferred to translational, rotational or vibrational states of the molecule. Through interactions with other molecules this rotational and vibrational energy can be converted into translational energy creating a temperature rise in the fluid. In our experiment, following

each pulse from the laser, the fluid is allowed to cool to an ambient temperature. This alternate heating and cooling creates a pressure variation which is observed as acoustic pulses.

Present optoacoustic theories<sup>4,5,6</sup> assume that the energy transfer processes discussed above all take place over a period of time which is very short compared to other factors that effect the observed pressure profile; such as, the length of the laser pulse and its diameter as it enters the fluid.

The process of energy transfer and the rate at which it occurs varies from one liquid to another because the energy levels and rate of energy transfer depend on the structure of the molecules. If the rate of production of translational energy is slow due to internal relaxation processes, the shape of the acoustic pulse should be governed by these relaxation processes. The experimental system must be capable of resolving very short acoustic pulses in order to resolve events occurring on this time scale. Otherwise, the pressure profile is dominated by geometrical factors of the experiment.

Carbon disulfide ( $\text{CS}_2$ ) has been chosen as one subject of study because previous ultrasonic absorption measurements<sup>7</sup> have already shown that the transfer of energy between translational and vibrational states is relatively slow (compared, for example, to  $\text{H}_2\text{O}$ ).

Mr. Charles Thompson completed his M.S. work in December of 1987 which was a study of energy transfer in  $\text{CS}_2$ . His Thesis was issued

as a Technical Report.<sup>8</sup>

## 1.2 Experimental Configuration

The experimental setup will be described here for completeness even though a similar description has been provided in previous reports.

A block description of the experimental technique is shown in Figure 1.1. This technique utilizes two lasers. A 1.25 MW nitrogen laser emitting 800 ps pulses in the UV (337nm) is used as the optoacoustic source. A 3mW continuous He-Ne laser runs parallel to the excitation zone. It is then focused on a photodiode which is coupled to a digitizing oscilloscope. The probe laser responds to refractive index gradients which are produced as a result of the acoustic wave. These gradients cause small deflections of the probe beam which are detected by the photodiode. The observed signal is proportional to the pressure gradient. Comparison to theory can be achieved by either integrating the observed signal or by differentiating the predicted pressure. Both techniques have been used; however, a comparison to the absolute magnitude of the acoustic pressure can be achieved as well if the theory is compared to the output of a PVDF hydrophone as shown in Figure 1.2. Given the inaccuracies in beam size estimates, the agreement between theory and experiment is considered acceptable.



Energy delivered to the fluid is varied by placing glass slides in the beam path of the UV laser. N number of glass slides (~.23 mm thick) are supported by two small pieces of plexiglas. The centers of the plexiglas have been removed. To provide for accurate energy measurements, twenty sets of attenuators corresponding to a different number of glass slides were made. The energy transmission for each attenuation was measured with an energy meter. The precision of the energy meter is  $\pm 1\mu\text{J}$ . Figure 1.3 shows measured energy delivered to the test cell as a function of the number of glass slides.

The energy density in the fluid can also be increased by increasing the optical absorption coefficient  $\alpha$ . This is achieved by either adding dye to the test liquid or by choosing a liquid that strongly absorbs UV radiation ( $\text{CS}_2$ ).

#### Determination of the Excitation Beam Size

The dimensions of the excitation beam are among the most important parameters involved in theoretical predictions. An accurate measurement of the spatial beam profile is vital for comparison to theory.

The Nitrogen laser used emits an 800 ps UV pulse, rectangular in shape. The ratio of the two sides of the rectangular beam can be determined by passing a small piece of photographic film through the

beam in the vicinity of the focal point. The size of the exposed portion of a film measured under a microscope was found to be  $150\text{ }\mu\text{m} \times 550\text{ }\mu\text{m}$ . Although the absolute dimensions will depend upon the shape of the beam wings and exposure time, the width to height ratio  $550/150$  should remain constant.

The time duration of the acoustic pulse is primarily determined by the width of the short dimension of the excitation pulse. By comparing an experimental waveform to theory one can estimate the small dimension of the rectangular pulse. For this measurement, it is important to use a liquid with a fast relaxation time to insure that the shape of the acoustic pulse is not effected by relaxation effects. Fluorescence may also cause problems in beam size measurements due to radiative transport outside the initial excitation zone.

Alcohols and water are fast relaxing fluids but are not good absorbers of UV radiation ( $337\text{ nm}$ ). One way to increase the absorption rate is to add dye to the fluid, however the dye presents a problem with fluorescence. The liquid chosen for the measurements presented here is a clean brand of Scotch. It is alcohol based, a reasonably good absorber, relaxes rapidly, and fluorescences only weakly. This fluid provided the shortest optoacoustic signal. The comparison of experimental and theoretical waveforms are shown in Figure 1.4. The radius required to match the waveforms was  $45\text{ }\mu\text{m}$ . This suggests a rectangular profile of  $90\text{ }\mu\text{m} \times 330\text{ }\mu\text{m}$ . Using this technique the values given should be accurate to within  $\pm 10\text{ }\mu\text{m}$ .

### 1.3 Microscopic Interpretation of Optoacoustics in Fluids

At the UV wavelengths employed in this experiment, the laser photons which are absorbed result in electronic excitation of the recipient molecules. This electronic energy is then rapidly converted to vibrational and translational energy which is manifested macroscopically as thermal expansion. We conclude that the electronic excitation does not lead directly to thermal expansion by comparison of the UV results to measurements using IR lasers. The IR lasers produce photons with insufficient energy to cause electronic excitation yet the efficiency at which the electromagnetic energy is converted to thermal expansion is identical to that for the UV measurements (at least in the linear regime). Since the observed thermal expansion depends only upon the total energy deposited in the fluid and not on the optical wavelength, we conclude that it is vibrational and/or translational modes which give rise to expansion.

Further insight into the critical energy flow path can be gained from Thompson's<sup>8</sup> results. Specifically, in the case of CS<sub>2</sub>, he found that following deposition of energy by the laser, there was a time lag for resultant thermal expansion of about 90 nsec which seemed to be associated with transfer of energy from an electronic (initial) state to translation (or vibration) as well as a time constant of 1.5 nsec for vibrational relaxation. These results indicate that when energy is added to the fluid, that energy must undergo internal conversion processes prior to causing thermal expansion. Further, since the

laser used has a pulse length of 0.8 nsec, essentially all the laser energy deposited resides in internal states at the end of the pulse.

The observation that deposited energy resides in excited states for relatively long time periods gives rise to some interesting consequences. Specifically, for a large energy flux, this raises the possibility of depositing photons in a grouping of molecules more rapidly than the energy is redistributed in the fluid.

Refer to Figure 1.5. Up to an input energy of about 80  $\mu\text{J}$ , the optoacoustic signal strength varies linearly with input energy. The slope of the curve is consistent with linear thermal expansion. At 80  $\mu\text{J}$ , there appears to be a plateau followed by a range of energies where the signal strength increases more rapidly. At about 160  $\mu\text{J}$ , there is another plateau followed by an even steeper rise in optoacoustic signal strength with input energy. When the input energy reaches 210  $\mu\text{J}$ , the slope begins to decrease. There is a further decrease at 320  $\mu\text{J}$  returning the slope to near its low signal value. It is interesting to note that at each of the two plateaus, the optoacoustic conversion efficiency (the slope of the curve) increases by approximately 50%.

Higashigaki, et al.<sup>9</sup> has carried out investigations of molecular reorientations of carbon disulphid molecules using a light scattering technique. The reorientation of the  $\text{CS}_2$  molecular axis involves displacements of other molecules. He concludes that the activation

energy required to reorient a CS<sub>2</sub> molecule is 1.58 kcal/mol. Considering the size of our excitation volume, we find that there are  $1.33 \times 10^{-8}$  moles of CS<sub>2</sub>. This gives an activation energy of  $2.1 \times 10^{-5}$  cal or 88  $\mu$ J. Note that this occurs very near the first plateau suggesting that these reorientations may contribute to the increase in optoacoustic conversion efficiency.

The theory from which the beam size parameters were deduced<sup>10</sup> assumes a Gaussian spatial profile. Although a radial profile is assumed there, for purposes of energy disposition calculation, we will adopt a rectangular Gaussian described by

$$I = I_0 e^{-x^2/x_0^2} e^{-y^2/y_0^2} e^{-\alpha z}$$

where  $x_0$  is estimated to be 90  $\mu$ ,  $y_0$  is estimated to be 330  $\mu$ , and  $1/\alpha$  has been measured to be 27  $\mu$ . From the temporal response of the optoacoustic waveform, we conclude that the energy contribution is substantially constrained to a volume  $x_0 \times y_0 \times 1/\alpha$ . Some of this region will absorb more energy, other parts less. We will comment on this fact later.

Based upon the above, as a starting point, we will assume that the beam has an effective cross section of 90  $\mu \times 330 \mu$  giving an area of  $3 \times 10^{-8} \text{ m}^2$ . We will further assume that the pulse length is 800 psec (manufacturer's specification) so the input intensity at the first plateau (80  $\mu$ J) is  $3.3 \times 10^{12} \text{ W/m}^2$ . In accordance with Beer's

Law,

$$I = I_0 e^{-\alpha z}$$

where  $I_0$  is the input intensity,  $\alpha$  is the absorption coefficient, and  $z$  is the depth into the fluid. Differentiating,

$$dI/dz = -\alpha I_0 e^{-\alpha z}$$

gives the energy absorbed/unit volume-unit time, a distance  $z$  into the test cell. At the point where the excitation beam intensity is down to  $1/e$  of the initial value,  $\alpha z = 1$  and  $dI/dz = -\alpha I_0/e$ . Taking  $\alpha = 3.7 \times 10^4 \text{ m}^{-1}$  (penetration depth of  $27 \text{ } \mu\text{m}$ ),  $dI/dz = 4.5 \times 10^{16} \text{ J/m}^3\text{-s}$  at  $z = 1/\alpha$  and  $1.2 \times 10^{17} \text{ J/m}^3\text{-s}$  at  $z = 0$ .

The results of this energy deposition can be viewed macroscopically and microscopically. Microscopically, the energy absorbed can be viewed in terms of the number of photons. Taking  $\lambda = 3.4 \times 10^{-7} \text{ m}$ ,  $E = hc/\lambda$  gives an energy of  $5.9 \times 10^{-19} \text{ J/photon}$  so the fluid is absorbing between .76 and  $2.1 \times 10^{35} \text{ photons/s-m}^3$ . For comparison, at  $20^\circ\text{C}$ , the density of  $\text{CS}_2$  is about  $1.3 \text{ g/cm}^3$ <sup>11</sup> and since each  $\text{CS}_2$  molecule has a mass of  $1.3 \times 10^{-22} \text{ g}$ , there are approximately  $10^{22} \text{ molecules/cm}^3$  or  $1.03 \times 10^{28} \text{ molecules/m}^3$ . If the excitation laser operated in the steady state, each molecule would absorb  $1\text{-}2 \times 10^7$  photons but since the pulse is only 800 psec in duration, each molecule absorbs only .02 photons during the laser

pulse. Another way to view this is that if the photon density exceeds about 0.02 photons/molecule, a plateau in optoacoustic conversion energy is reached -- something different begins to happen.

Next, we will adopt a macroscopic view. In the defined excitation volume, the energy absorbed near the first plateau is about  $1.0 \times 10^8 \text{ J/m}^3$ . The specific heat of  $\text{CS}_2$  liquid near room temperature is  $76 \text{ J/g-mol-K}^{12}$ . The density is  $1.3 \text{ g/cm}^3$  or  $0.017 \text{ g-mol/cm}^3$  so  $C_p(l)$  is  $1.3 \times 10^6 \text{ J/m}^3\text{-K}$  resulting in a temperature rise (ignoring temperature dependence of density and  $C_p(l)$ ) of  $77^\circ\text{K}$ . The boiling point of  $\text{CS}_2$  is  $46^\circ\text{C}$ . Assuming a starting temperature of  $23^\circ\text{C}$ , the plateau at  $80 \mu\text{J}$  is in excess of the energy necessary to raise the fluid to its boiling point throughout the focal volume.

Two minor details should be noted prior to continuing this argument. First, the excitation laser intensity is greatest next to the quartz surface at the beam entry location. The fluid is in contact with a surface which should prohibit supersaturation. Second, microbubbles are not observed until the input energy reaches about  $120 \mu\text{J}$ . At an energy required to produce bubbles, no unusual dependence of optoacoustic signal strength with input energy is observed.

The heat of vaporization of  $\text{CS}_2$  is  $89.56 \text{ g cal/g}$  or  $4.9^{13} \times 10^8 \text{ J/m}^3$ . The first  $2.7 \times 10^7 \text{ J/m}^3$  is required to reach the boiling point (prior to the  $80 \mu\text{J}$  plateau). The next  $4.9 \times 10^8 \text{ J/m}^3$  is

required to provide the heat of vaporization at  $z = 0$ , the total energy required to vaporize the fluid would be  $400 \mu\text{J}$ . Note, however, that between the  $80 \mu\text{J}$  and  $160 \mu\text{J}$  plateaus small bubbles begin to appear and the slope of the curve increases. This suggests that in the focal volume where the laser intensity is at its highest, local vaporization occurs at lower energies

From a macroscopic point of view, the signal strength versus input energy is now clear at least up to the point near  $210 \mu\text{J}$  where the slope begins to decrease. Specifically, at low input energies, the optoacoustic amplitude increases linearly with input energy up to  $80 \mu\text{J}$ . At this point, sufficient energy is deposited to bring the fluid to the boiling point. Further increases in input energy serve only to provide heat of vaporization. Once some of the fluid is vaporized, the coefficient of thermal expansion becomes dominated by those gas phase molecules present. The gas phase has an expansion coefficient (assuming an ideal gas) of  $3.1 \times 10^{-3}/\text{K}$  while the liquid phase has an expansion coefficient of  $1.2 \times 10^{-3}/\text{K}$ . Referring to Figure 1.5, the slope of the curve at low energy is, indeed, 0.4 times that above  $180 \mu\text{J}$ . Between these two slopes, there is an intermediate region where part of the energy is being used to do expansion work but most is going into the heat of vaporization.

Now that the general features of the experimental observation have been explained from a macroscopic viewpoint, again consider what is happening at the molecular level. To begin this description, one more important experimental observation needs to be noted.



Specifically, as the energy deposited in the fluid increases, the optoacoustic waveform does not change in shape. Since the shape is controlled by internal relaxation processes, this observation proves that energy must appear in translation and vibration prior to an observable expansion or change in state. We can and will assume, therefore, that all processes at the molecular level start after energy is transferred to v-t states.

We will view the liquid as a dense gas or imperfect lattice characterized by some radial distribution function  $g(R)$  which gives the probability of finding a second molecule a distance  $R$  from a reference molecule. For a perfect lattice,  $g(R)$  would be periodic along any axis; for a gas at low density, it would be zero out to the molecular diameter and uniform beyond. Since we envision the liquid as an intermediate state,  $g(R)$  should be a damped periodic function which decays toward a constant value more rapidly with increasing temperature until, at the boiling point,  $g(R)$  approaches the gas phase value. Indeed, microscopic description of the liquid state typically involves expressions for  $g(R)$  in terms of intermolecular potentials.

#### Equation of State

We have shown that for large deposition energies, the liquid in the excitation volume may undergo a change in phase. Heating of the volume will change the thermodynamic properties of the fluid which

enter into subsequent calculation.

An equation relating the volume, temperature, and other thermodynamic properties needs to be found. This will allow a comparison of experimental data to theoretical predictions.

Experimental data of differential volume as a function of temperature is available for carbon disulphide.<sup>14</sup> The equation of state used for this comparison is Van der Waals' equation expanded out to four terms.<sup>15</sup> It is given as

$$P + \frac{a}{V^2} = \frac{RT}{V} \left[ 1 + \frac{b}{V} + .625 \left( \frac{b}{V} \right)^2 + .2869 \left( \frac{b}{V} \right)^3 + .1928 \left( \frac{b}{V} \right)^4 \right].$$

Temperature as a function of volume was plotted and is shown in Figure 1.6. The constants  $a$  and  $b$  in the operating state were adjusted to fit this data. The adjusted values of  $a$  and  $b$  are  $41.6 \text{ L}^2 \cdot \text{atm/mol}^2$  and  $.173 \text{ L/mol}$ , respectively. The accepted values of  $a$  and  $b$  for  $\text{CS}_2$  gas are given as  $11.62 \text{ L}^2 \cdot \text{atm/mol}^2$  and  $.07685 \text{ L/mol}$ , respectively.<sup>16</sup> Our values should be considered to be empirical parameters only.

# Experimental Configuration

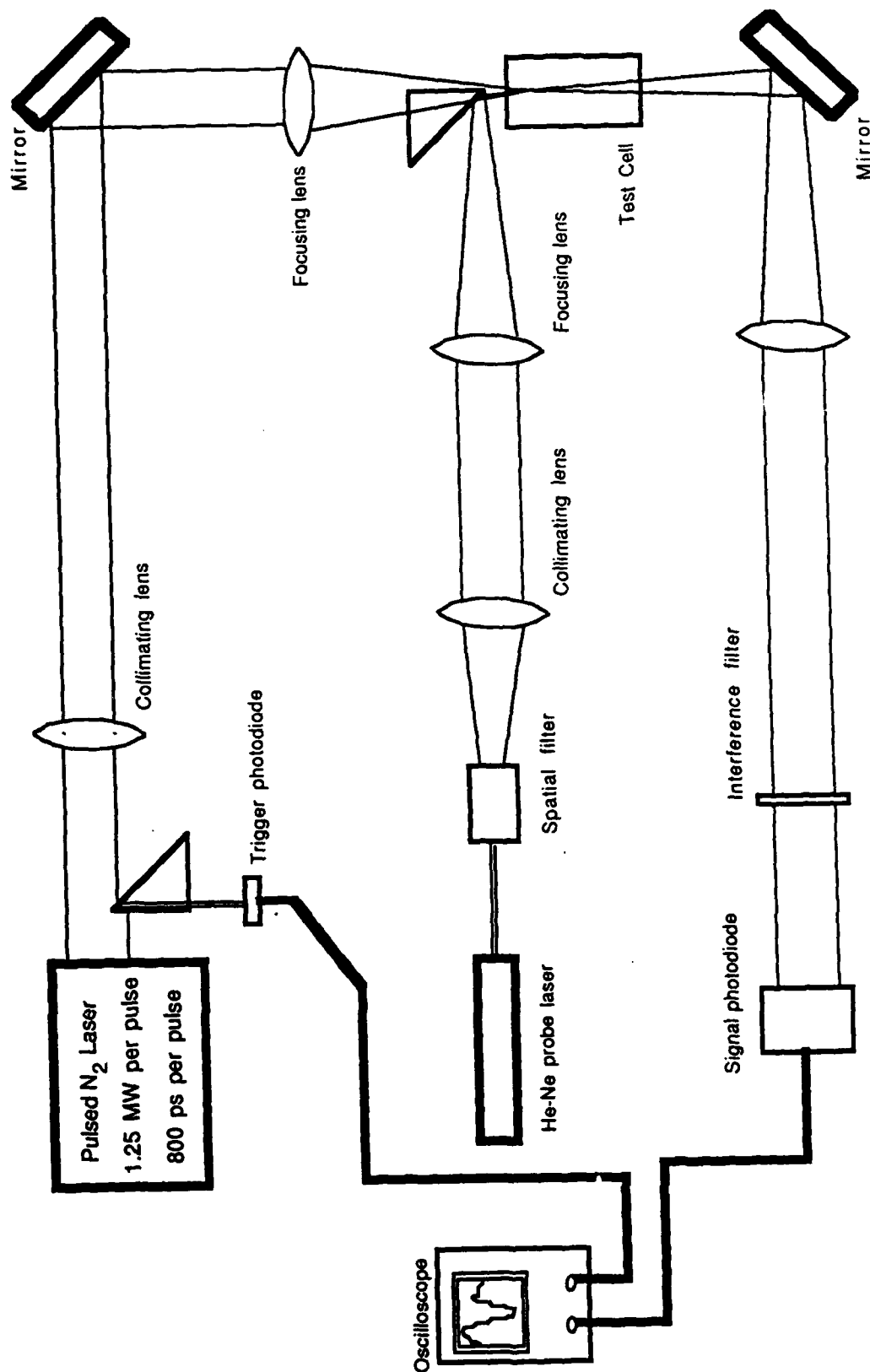


Figure 1

A vs PT

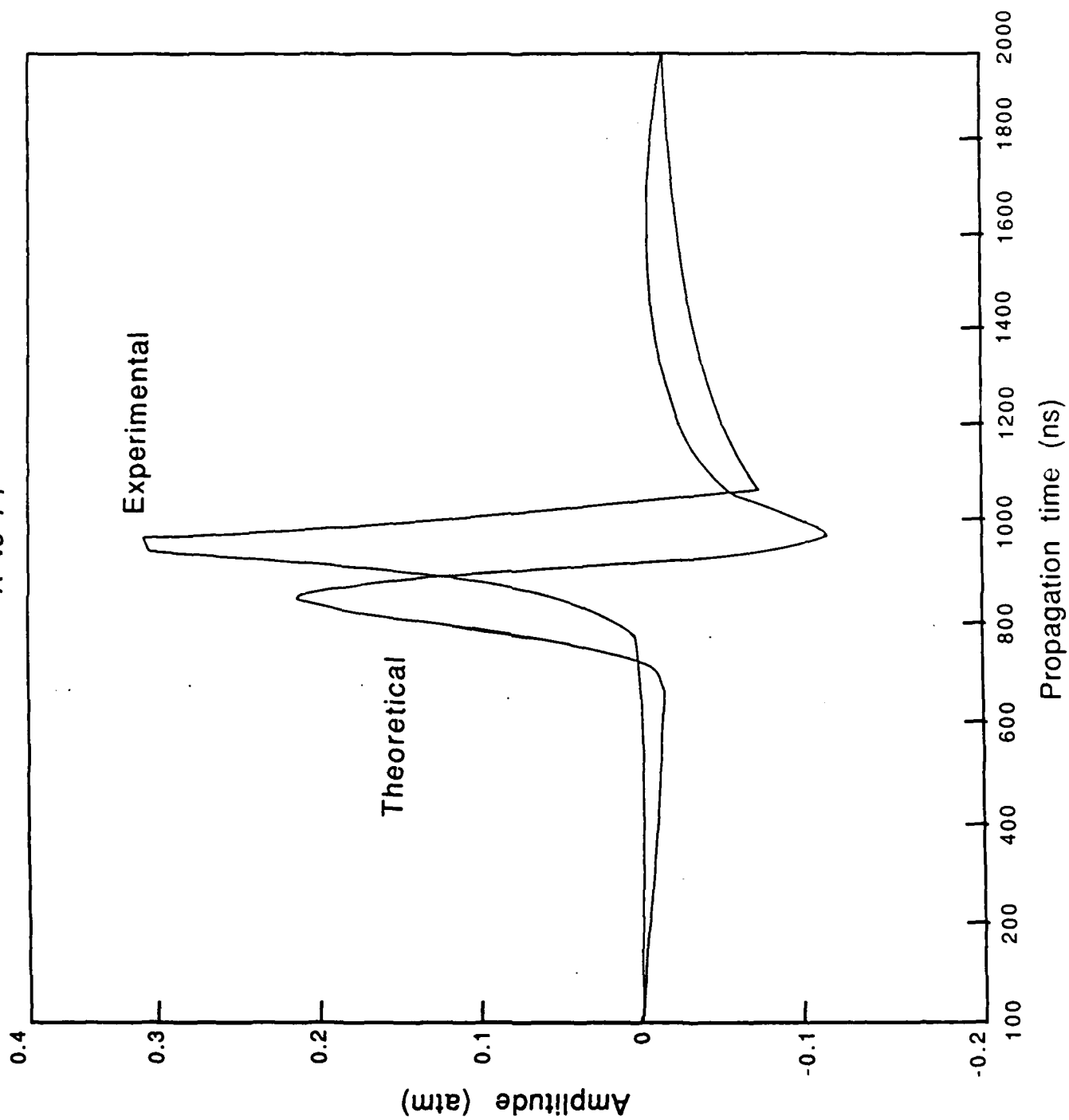
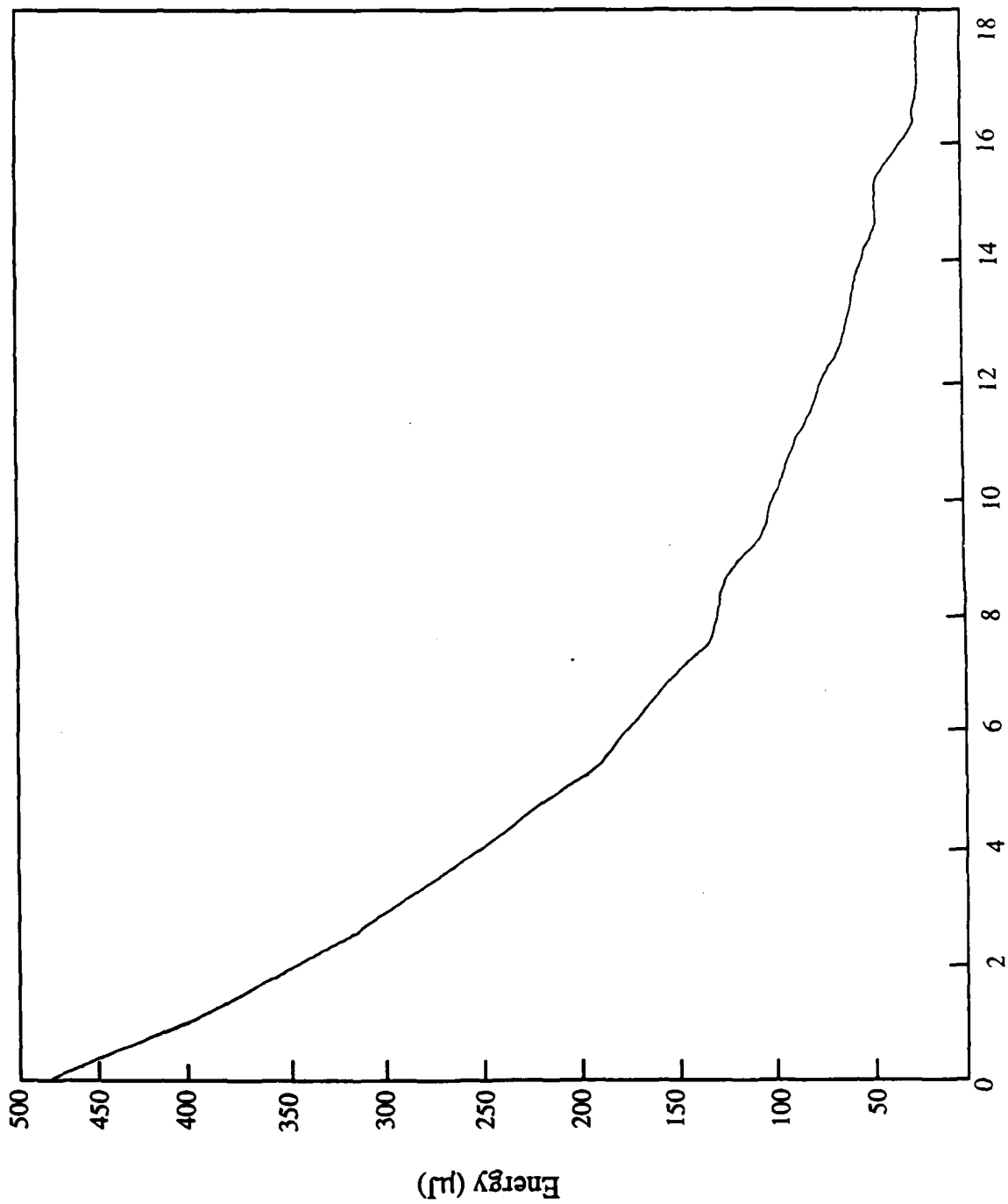


Figure 1.2

# Energy vs Number of Slides



Number of Slides (.23 mm/slide).

Figure 1.3

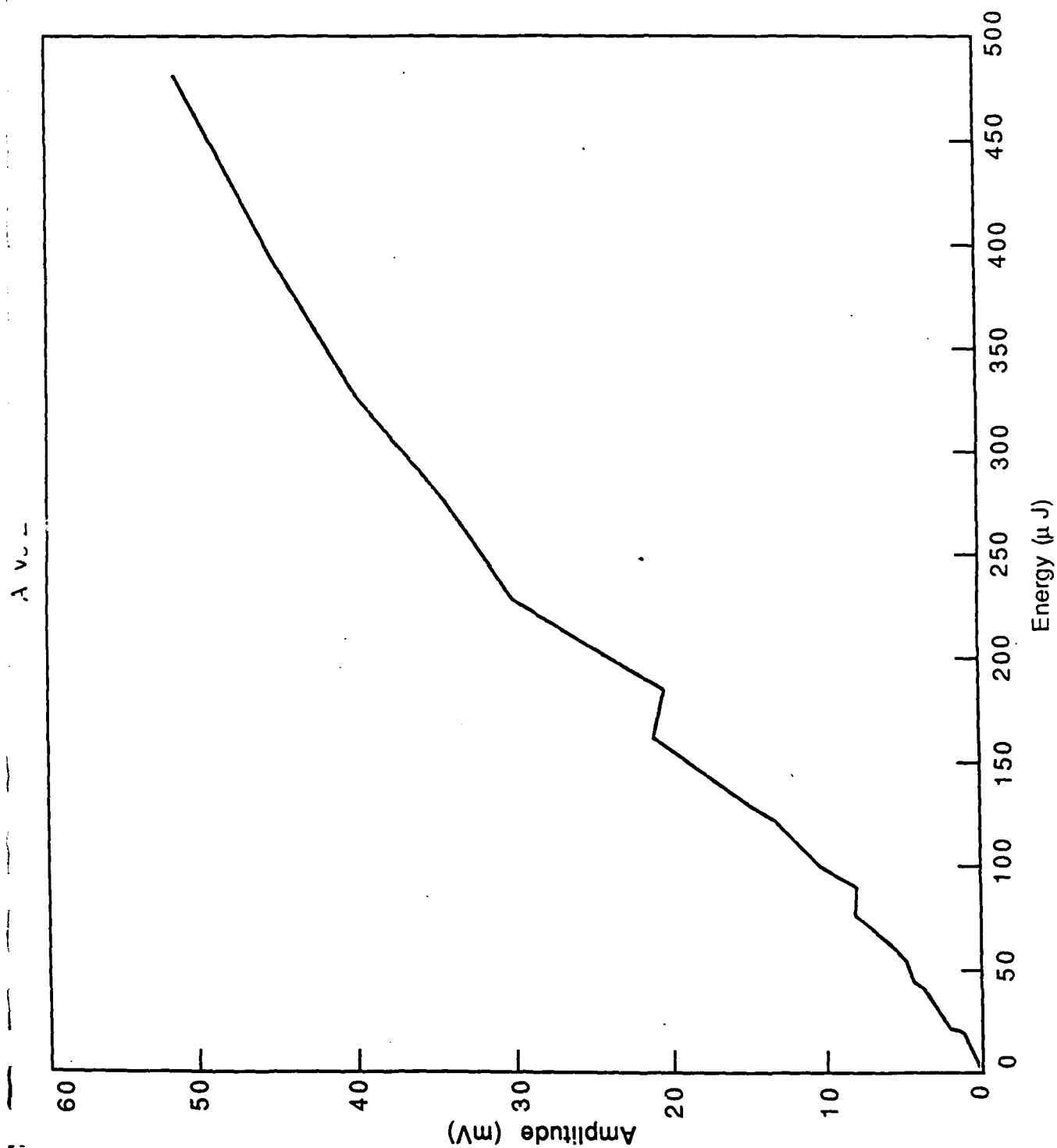


Figure 1.4

# Beam Width Determination

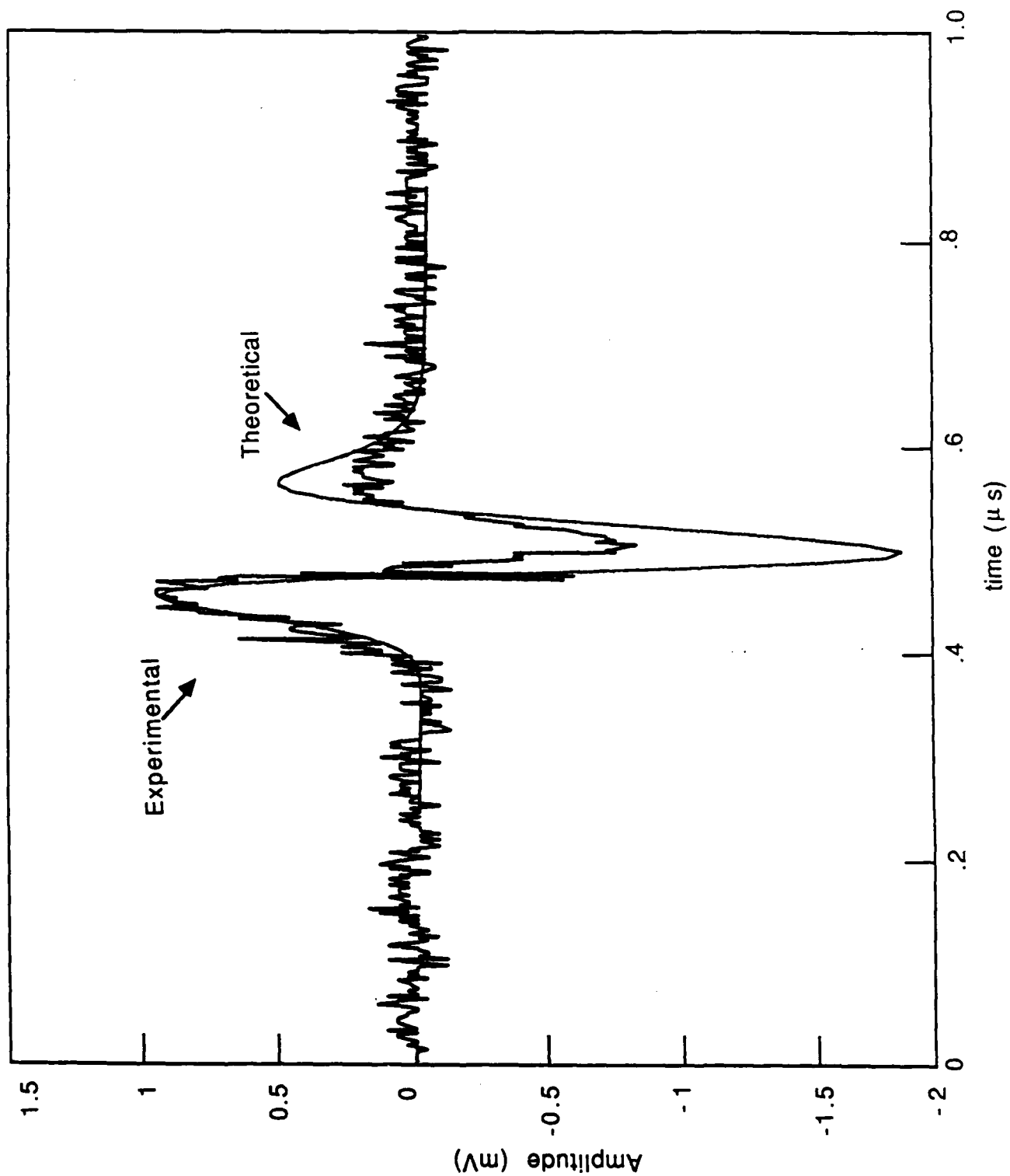


Figure 1.5

Experiment vs Theory

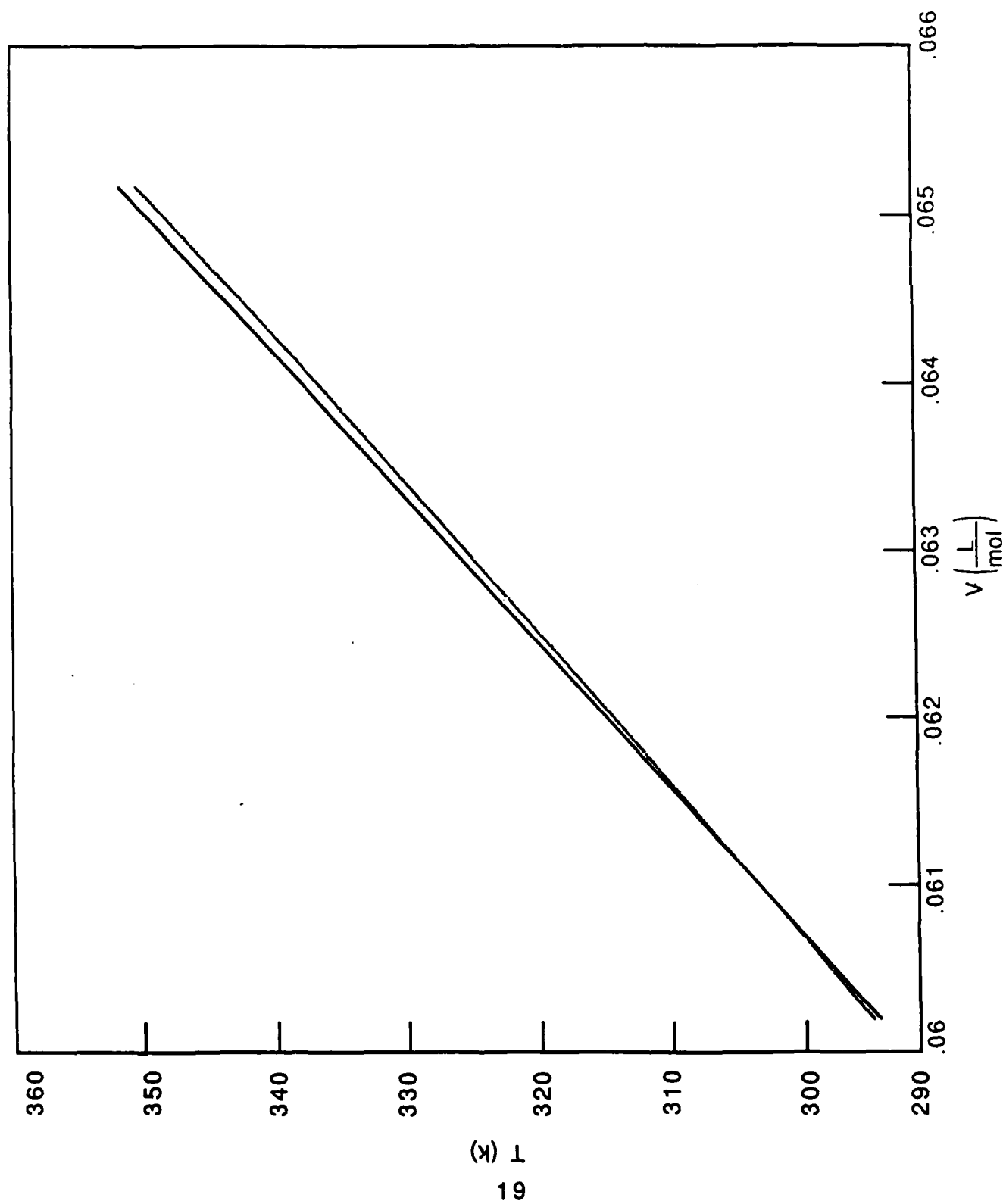


Figure 1.6



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## 2.0 Ultrasonic Attenuation in Polymer/Water Mixtures

During the previous one year period, this lower prior task has not been actively pursued due to difficulties in obtaining polymer samples of interest.

### 3.0 Propagation of Sound in Vibrationally Excited Gases (SACER)

#### 3.1 Summary of Past Work

This project was begun several years ago as a search for SACER (Sound Amplification from Controlled Excitation Reactions). The effect has been observed in  $N_2$  with added amounts of He,  $H_2$ ,  $H_2O$  and  $CH_4$  and the measured amplification found to exceed theoretically predicted values. In the process, the experimental procedure has enabled the measurement of some here-to-fore difficult-to-observe relaxation times and their temperature dependence. These results were reported in a paper entitled "Propagation of Sound in Vibrationally Excited  $N_2/He$ ,  $N_2/H_2$ ,  $N_2/CH_4$ , and  $N_2/H_2O$  Mixtures" published in JASA 83, 2186 (1988).

The experimental method has been described in earlier reports. It involves exciting the vibrational states in a gas with an electrical discharge and observing the change in velocity and absorption of sound in the gas over a period of a few tens of milliseconds following the discharge. In  $N_2$  and a few other selected gases most of the electrical discharge energy winds up in molecular vibrational energy in a few microseconds. By measuring the energy the discharge dumps into the gas, we have estimated the vibrational temperature immediately following the discharge. Vibrational temperatures 1000 to 2000 K above translational temperatures are obtained in this way.

Translational temperatures are determined from the sound velocity which, in turn, is determined from the resonant frequency of the discharge tube which changes with time following the discharge. The sound velocity in this way becomes a thermometer able to sense the gas temperature changes on a millisecond time scale, and the discharge tube a kind of spectraphone that measures the rate of conversion of vibrational energy into translational energy.

### 3.2 Work During the Past Year

We have spent the past year working primarily with carbon monoxide. This gas was chosen because calculations from electron collision cross sections showed that it could also be excited vibrationally by an electrical discharge.<sup>1</sup> It was expected to behave in a way similar to  $N_2$  but to have the additional property of being infrared active. We, therefore, expected to be able to observe the decay of the vibrational temperature directly from the emitted infrared. (In  $N_2$  this decay was only inferred from the resulting rise and fall of the translational temperature). Tim Ruppel is working on this problem for his Master's Thesis. Efforts to date can be summarized under three headings:

- 1) Bibliography of previous work with CO,
- 2) Efforts at vibrationally exciting CO with an electrical discharge,
- 3) Measurement of vibrational temperatures in  $N_2$  using CO as

the monitoring gas.

These three areas are discussed separately below.

### 3.2a Bibliography of Previous Work with CO

#### Papers Concerning Electronic Excitation in CO Discharges

1. P.A. Sergeev and D.I. Slovetsky, J. Chem. Phys. 75, (231-41) 1975.

#### Papers Concerning VT transfer in pure CO

1. M.A. Kovacs and M.E. Mack, Appl. Phys. Lett. 20, (487-90) 1973.
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4. John C. Stephenson, Appl. Phys. Lett. 22, (576-81) 1973.

#### Papers Concerning VT transfer in CO mixtures

1. John C. Stephenson, Appl. Phys. Lett. 22, (576-8) 1973.
2. Richard G. Miller and J.K. Hancock, J. Chem. Phys. 66, (5150-8) 1977.
3. Donald J. Miller and Roger C. Millikan, J. Chem. Phys. 53, (3384-5) 1970.
4. Roger C. Millikan, J. Chem. Phys. 38, (2855-60) 1963.
5. C. W. von Rosenberg, Jrl, K.N.C. Bray, and N.H. Pratt, J. Chem. Phys. 56, (3230-7) 1972.

6. Roger C. Millikan and Donald R. White, J. Chem. Phys. 39, (98-101) 1963.
7. John C. Stephenson and Earl R. Mosburg Jr., J. Chem. Phys. 60, (3562-6) 1974.
8. D.F. Starr, J.K. Hancock and W.H. Green, J. Chem. Phys. 61 (5421-5) 1974.
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10. William J. Hooker and Roger C. Millikan, J. Chem. Phys. 38, (214) 1963.

#### **Papers Concerning VV Transfers in Pure CO**

1. John C. Stephenson, Appl. Phys. Lett. 22, (576-8) 1973.
2. Mario Cacciatore and Gert Due Billing, J. Chem. Phys. 58, (395-407) 1981.
3. John C. Stephenson and Earl R. Mosburg Jr., J. Chem. Phys. 60, (3562-6) 1974.

#### **Papers concerning VV transfers in CO mixtures**

1. Roger G. Miller and J.K. Hancock, J. Chem. Phys. 66, (5150-8) 1977.
2. D.C. Allen and C.J.S.M. Simpson, J. Chem. Phys. 45, (203-11) 1980.
3. E.T. Chandler, E.A. Gregory, R.M. Siddles and C.J.S.M. Simpson, J. Chem. Phys. Lett. 39, (347-9) 1976.
4. G. Mastrocinque, A. Chakroun, L. Doyennette, H. Gueguen, M. Margottin-Maclou and L. Henry, J. Chem. Phys. Lett. 39, (347-9) 1976.

5. C.W. von Rosenberg Jr., K.N.C. Bray and N.H. Pratt, J. Chem. Phys. 56, (3230-7) 1972.
6. John C. Stephenson and Earl R. Mosburg Jr., J. Chem. Phys. 60, (3562-6) 1974.
7. D.F. Starr, J.K. Hancock and W.H. Green, J. Chem. Phys. 61, (5421-5) 1974.
8. Paul F. Zittel and C. Bradley Moore, Appl. Phys. Lett. 21, (81-3) 1972.
9. W.H. Green and J.K. Hancock, J. Chem. Phys. 59, (4326-35) 1973.
10. William H. Green and J. Kent Hancock, IEEE Journ. Quan. Elec. QE-9, (50-8) 1973.

**Papers Concerning the Radiative Lifetime of CO**

1. Roger C. Millikan, J. Chem. Phys. 38, (2855-60) 1963.

**Papers Concerning our Experimental Determination of N<sub>2</sub> Temperatures.**

1. Esmail Jamshidi and Ralph H. Kummeler, J. Chem. Soc. Faraday Trans. II, 75, (1205-7) 1979.
2. D.C. Allen, E. T. Chandler, E. A. Gregory, R. M. Siddles and C.J.S.M. Simpson, J. Chem. Phys. Lett. 76, (347-53) 1980.



### 3.2b. Efforts at Vibrationally Exciting CO in an Electrical Discharge

So far we have been successful in getting only small amounts of energy into CO and CO light gas mixtures with our electrical discharge. This is evidently because of the larger conductivity of the CO gas. We are continuing an effort to find a discharge mechanism that give us greater vibrational excitation of CO.

### 3.2c. Measurement of the Vibrational Temperature of $N_2$ Using CO as a Monitoring Gas

Perhaps the most significant accomplishment of the past year has been the successful measurement of vibrational temperatures in  $N_2$  using CO as an indicator gas. The vibrational energy levels in CO and  $N_2$  are sufficiently close so that, when a trace amount of CO is mixed with  $N_2$ , the CO vibrational energy will maintain thermal equilibrium with the  $N_2$  vibration and the two vibrational temperatures decay together.<sup>2</sup> The infrared activity of CO can therefore be used to follow the vibrational energy of  $N_2$  even through the non-polar  $N_2$  molecule does not radiate.

The vibrational temperatures have been measured using a cold gas cell method.<sup>3</sup> For this purpose, the electrical discharge tube has been terminated with a cylindrical cell 7.31 cm long with CaF end windows. (See Fig.3.1) When CO gas at atmospheric pressure is introduced into this cell at room temperature, the part of the

infrared corresponding to the 1 to 0 vibrational transition will be absorbed. The ratio of the infrared intensity coming through the empty cell to that coming through the full cell should be given by the equation <sup>5</sup>

$$\frac{I(\text{empty}) - I(\text{full})}{I(\text{empty})} = (1 + \epsilon)^{-2}$$

$$\text{Where } \epsilon = \left[ \exp \left( \frac{h \omega}{k T_v} \right) - 1 \right]^{-1}$$

This equation therefore, can be solved to give the vibrational temperature.

Figure 3.2 shows results of measurements on a mixture of N<sub>2</sub> and CO. Curve 1 is the translational temperature as determined from the sound velocity measurements. The translational temperature changes little with time due to the slow v-t rate in pure N<sub>2</sub>. Curve 2 is the vibrational temperature of N<sub>2</sub> calculated from the translational temperature assuming a relaxation model and known values for the thermal conduction and vibrational relaxation times as explained in ref.4. Curve 3 is the vibrational temperature of CO as determined from the decay of the CO infrared radiation using the "cold-gas-cell" method described above. The difference between curves 2 and 3 caused us no little concern until we remembered that even though the vibrational modes in N<sub>2</sub> and CO are in equilibrium, the principle of detailed balancing predicts that their vibrational temperatures will not be equal if the vibrational temperatures differ from the translational temperature. The relationship between the vibrational

temperature of  $N_2$  ( $T_{N_2}$ ), the vibrational temperature of CO ( $T_{CO}$ ), and the translational temperature ( $T$ ) is given by<sup>3</sup>

$$\frac{V_{CO} - V_{N_2}}{T} = \frac{V_{CO}}{T_{CO}} - \frac{V_{N_2}}{T_{N_2}},$$

where  $V_{CO}$  and  $V_{N_2}$  are the fundamental vibrational frequencies of the CO and  $N_2$  molecules using this equation, curve 4 in Fig.3.2 is calculated from curves 2 and 1. This agreement between the measured relaxation of the vibration temperature (curve 4) and that inferred from the energy deposited in the gas by the electrical discharge (curve 2) is very gratifying and gives us confidence in our method of determining relaxation times and their temperature dependence.

### 3.3 Work Planned for the Coming Year

The discovery that we can use CO as an "indicator gas" to measure the decay of vibrational temperatures in our SACER-SPECTRAPHONE opens up some interesting possibilities. One that we want to pursue during the coming year is the measurement of the vibrational relaxation of  $H_2$ . The very large spacing between the vibrational energy levels in  $H_2$  means that  $H_2$  gas must be at several thousand degrees before the gas contains appreciable vibrational energy. For that reason no one has ever measured vibrational relaxation rates in  $H_2$ . Electron collision cross section values indicate that  $H_2$  like  $N_2$  and CO should be vibrationally excited by an electric

discharge. In addition, the second vibrational level in CO is matched almost exactly with the first level in H<sub>2</sub>. Therefore v-v exchange between CO and H<sub>2</sub> should be fast and we should be able to use CO as an "indicator gas" to follow the decay of vibrational energy in H<sub>2</sub> as was discussed above for N<sub>2</sub>. If this is possible the variation of the relaxation time in H<sub>2</sub>/CO mixtures with CO concentration should give some valuable information not previously available about v-t and v-v rates in this and other H<sub>2</sub> gas mixtures.

# Cold Gas Filter

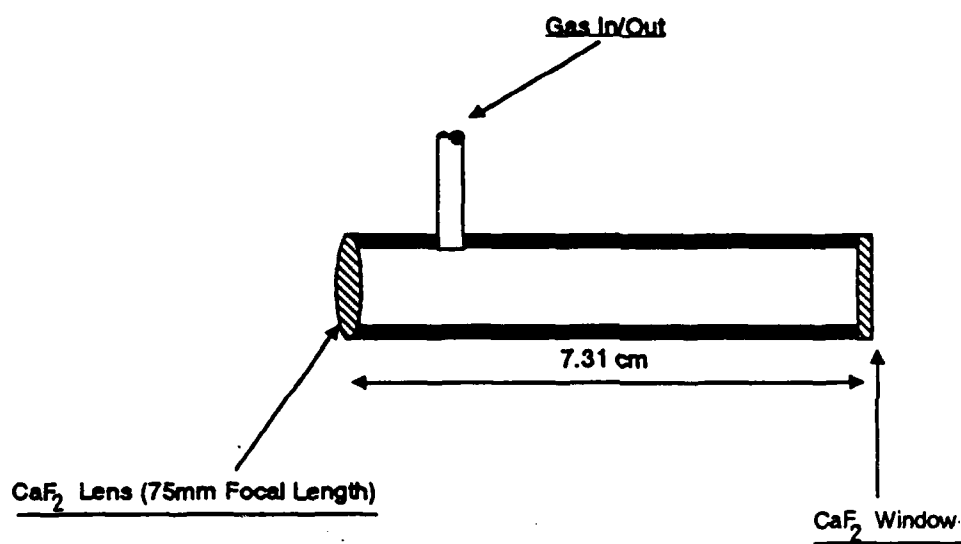


Fig. 3.1

# Vibrational & Translational Temperature of Nitrogen/Carbon Monoxide Mixtures

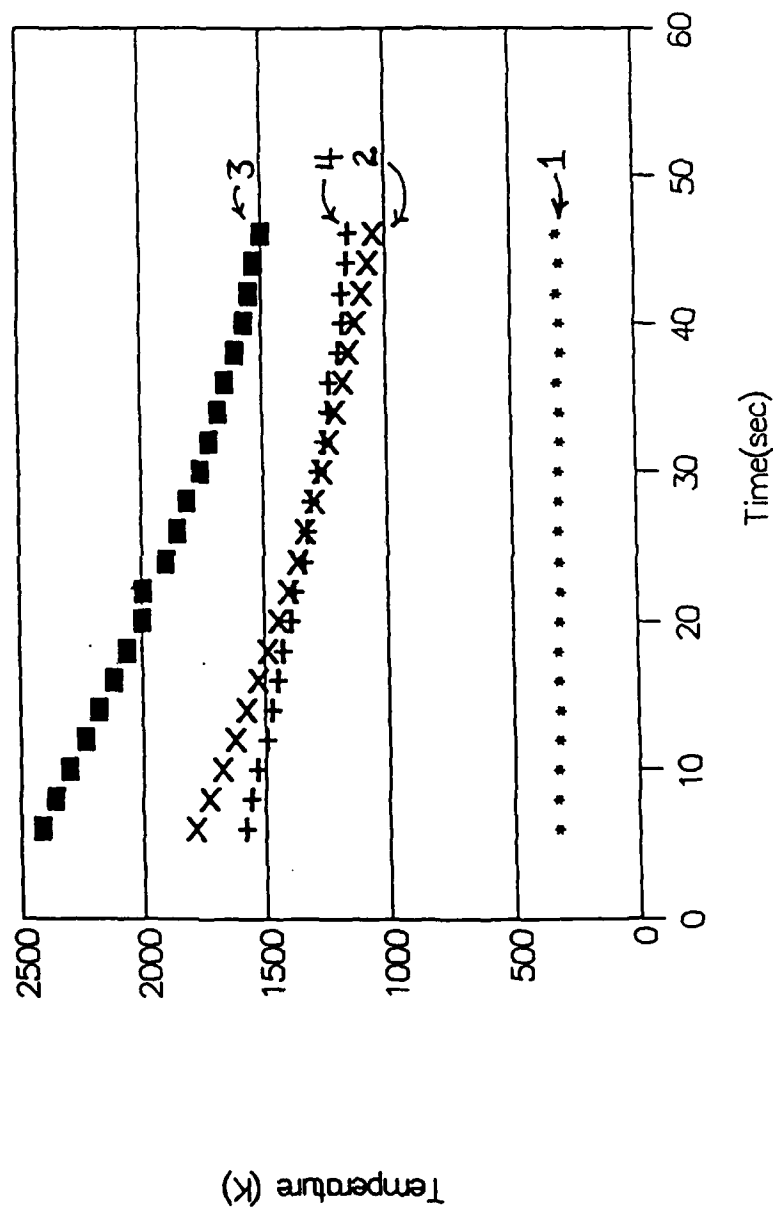


Fig. 3.2

## References

1. W. L. Nigham, Phys. Rev. A2, (1989-2000) 1970, App. Phys. Lett 20, (96-99) 1972.
2. E. Jamshidi and R. H. Kummier, J. Chem. Soc. Faraday Trans. II, 75, (1205-7) 1979.
3. E. Weitz and G. Flynn, Adv. Chem. Phys. (185-235) 1981 Eq. 40.
4. F. Douglas Shields and L. Dwyann Lafleur, J. Accus. Soc. Am. 83, (2186) 1988.
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June 1988

OFFICE OF NAVAL RESEARCH  
PUBLICATIONS / PATENTS / PRESENTATIONS / HONORS REPORT  
FOR

1 OCTOBER 19 87 through 30 SEPTEMBER 19 88

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CONTRACT NO0014 - 84-C-0193

R&T NO. 4126-936

TITLE OF CONTRACT: A Continued Study of Optical Sound Generation and Amplification

NAME(S) OF PRINCIPAL INVESTIGATOR(S) Henry E. Bass and F. Douglas Shields

NAME OF ORGANIZATION: Department of Physics and Astronomy

ADDRESS OF ORGANIZATION: University, MS 38677

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Page 1 of Enclosure (3)



June 1988

PAPERS SUBMITTED TO REFEREED JOURNALS  
(Not yet published)

C. H. Thompson, S. A. Cheyne, H. E. Bass, and R. Raspet, "The Effect of Internal Relaxation on Optoacoustic Conversion in Liquids". Submitted to J. Acoust. Soc. Am. (1988).

PAPERS PUBLISHED IN REFEREED JOURNALS

F. Douglas Shields and L. Dwyann Lafleur, "Propagation of sound in vibrationally excited  $N_2/He$ ,  $N_2/H_2$ ,  $N_2/CH_4$ , and  $N_2/H_2O$  mixtures", J. Acoust.Soc. Am. 83, 2186-2189 (1988).

June 1988

PAPERS PUBLISHED IN NON-REFEREED JOURNALS

N/A

TECHNICAL REPORTS PUBLISHED

Henry E. Bass and F. Douglas Shields, "A Continued Study of Optical Sound Generation and Amplification", PARGUM 87-4, Physical Acoustics Research Laboratory, The University of Mississippi, 38677 (1987).

Charles Thompson and Henry E. Bass, "The Effect of Internal Relaxation on Optoacoustic Conversion in Liquids", PARGUM 88-01, Physical Acoustics Research Laboratory, The University of Mississippi, 38677 (1988).

June 1988

BOOKS (AND SECTIONS THEREOF) SUBMITTED FOR PUBLICATION

N/A

BOOKS (AND SECTIONS THEREOF) PUBLISHED

N/A

June 1988

PATENTS FILED

N/A

PATENTS GRANTED

N/A

June 1988

INVITED PRESENTATIONS AT TOPICAL OR  
SCIENTIFIC/TECHNICAL SOCIETY CONFERENCES

N/A

HONORS/AWARDS/PRIZES

Henry E. Bass was appointed F. A. P. Barnard Distinguished Professor in May 1988.

Henry E. Bass was selected as UM HEADWAY Educator of the Year, 1988.

June 1988

GRADUATE STUDENTS SUPPORTED UNDER  
CONTRACT FOR YEAR ENDING 30 SEPTEMBER 19 88

Stanley Cheyne	(Ph.D Candidate)
Charles Thompson	(Received M.S. in December, 1987)
James Belcher	(M.S. candidate)
Jinxing Tan	(M.S. candidate)
Tim Ruppel	(M.S. candidate)

POSTDOCTORALS SUPPORTED UNDER  
CONTRACT FOR YEAR ENDING 30 SEPTEMBER 19\_\_

N/A



A CONTINUED STUDY OF OPTICAL  
SOUND GENERATION AND AMPLIFICATION

by

HENRY E. BASS AND F. DOUGLAS SHIELDS

STATUS REPORT

Graduate Students

Tim Ruppel, Ph.D. student in physics  
Stan Cheyne, Ph.D. student in physics  
Janxing Tan, Ph.D. student in physics  
James Belcher, M.S. student in physics

Graduates in the Past Year

Charles Thompson, M.S. in physics, now working for NORDA

Other Government Sponsored Research

Henry E. Bass is Principle Investigator for a USDA grant for construction of the National Center for Physical Acoustics (\$9.4M). Expected completion is January, 1990.

Status of Contract Funds

As of 30 October, \$13,222.67 in contract funds remain. Of this sum, \$7,457 is designated for equipment. We would like to carry approximately \$5,000 of these equipment funds into 1989. Little if any of the remaining funds will remain at the end of this contract period. In addition to contract funds, \$19,956.93 remain in UM matching funds. Of this, \$14,500 has been committed for a new data acquisition system and the remainder will be carried into 1989.

Permanent Equipment Purchased

To date, the only item purchased from contract funds has been an HP 5447D Wide Band Amplifier (~\$2,000). A new digital oscilloscope (~\$2,200) is on order but not yet received. This item should arrive prior to the end of this contract period. In addition to items purchased from contract funds, a Molection JD 1000 Joule Meter and Detector head were purchased from UM matching funds.