

FORCED RAYLEIGH SCATTERING IN LIQUIDS

Thesis for the Degree of M. S. MICHIGAN STATE UNIVERSITY VERENCE D. MOORE 1977

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ABSTRACT

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Forced Rayleigh Scattering was used to determine the thermal diffusivity of a water-polymer solution. We found we could get consistent results to within 1% for fixed d, with our equipment. Solutions of up to 5% by weight of polymer in pure water, had the same thermal diffusivity of pure water. When we used methyl-red to color our solution, we observe two times. One time $\ensuremath{\tau_s}$ which was due to the thermal diffusivity. The second time $\ensuremath{\tau_1}$ was due to some property of methyl-red in a basic solution.

FORCED RAYLEIGH SCATTERING IN LIQUIDS

By Verence D. Moore

A THESIS

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of Michigan State University in Partial
Fulfillment of the Requirements for
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MASTER OF SCIENCE
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I. INTRODUCTION

While Dr. Cowen was in France on Sabbatical, he was introduced to the technique of using Forced Rayleigh Scattering to determine the thermal diffusivity of liquid. The purpose of me doing this experiment was twofold. First, to test the apparatus and furnace to see if they were working and also to estimate the accuracy of the measurements. Secondly, to determine if we could see any effect of rather large amounts (up to 5% by weight of a high molecular weight polymer on the thermal diffusivity of water-polymer solutions.

The reason we only went up to 5% is because for solutions with greater amounts of polymer the solution can be considered a solid and we couldn't be able to get the solution into the cell. The polymer is Dow Separan AP30-high molecular weight ($\sim 10^6$) polyacrylamide. The interesting property of this polymer is that when it is mixed with water the solution becomes a gel. The gel's viscosity varies directly as the percent by weight of the polymer in the solution.

The way the solutions were made up was that we weighed out an amount of water. Then we weighed the approximate amount of polymer to correspond to 1%, 2.5% or 5% by weight of polymer to water. When we first started we put the polymer in first then added the water, but this caused a mixing problem. At the polymer water interface there was a high concentration solution, with

almost pure water and polymer on either side. By heating the samples for several hours (24 to 72), we were able to get a uniform solution. Later I found that I could get a uniform solution in about 16 to 32 hours by putting in half the water first, followed by the polymer, then the rest of the water.

The solution is normally clear so we had to add a dye to color the solution. The dye we used was methylred. The dye didn't readily mix with the solution, so heating the solution also help dissolve the dye.

After we had a uniform mixture we filtered the solution, to get rid of large particles of dye. We use five micron filter paper when filtering the solution. We use a millipore filter with a syringe to filter the solution. The syringe was used to force the solution through the filter system. We then put the filtered solution into our cells.

II. THE EXPERIMENT

A. Introduction

In this experiment we are using forced Rayleigh scattering to determine the thermal diffusivity of liquids. This is done by using two lasers of different wavelengths. One wavelength is transmitted by the liquid. The other wavelength is absorbed by the liquid. absorption of light is used to heat the sample in a small The heating laser beam is pulsed, heating the region. sample while the laser is on, and allowing it to cool while the laser is off. The heating beam, as it comes from the laser, is sent through two lenses which focus the beam. This allows us to choose the laser beam size inside the The beam size we use is about one millimeter in sample. diameter. Once the beam passes through the lenses it goes to a beam splitter. The beam splitter divides the beam into two equal intensity beams. The beam splitter consists of two mirrors; one fifty percent reflecting and the other one hundred percent reflecting. As the beam passes through the first mirror, fifty percent of the beam is reflected and fifty percent is transmitted. When the transmitted beam gets to the second mirror, one hundred percent of the beam is reflected. The beam splitter is positioned so that the two beams intersect inside of the sample. When the two beams intersect they form an interference pattern. This interference pattern causes a temperature grating in the region where the two beams meet.

Because of this the beam intensity in this region can be approximated as $I = I_0 \cos ky$, where I_0 is the maximum beam intensity inside the sample, and $k = \frac{2\pi}{d}$ where d is the distance between fringes.

If we superimpose the second laser beam, which is transmitted by the liquid, over the two intersecting heating beams the transmitted laser beam sees a diffraction grating. This grating is caused by the heat grating Because there is a temperature grating, the index of refraction is different for different areas inside this region where the heating is going on. Therefore, the effective path length the light must travel is different for different areas. Since the transmitted beam sees a diffraction grating as it passes through the sample, it forms a diffraction pattern after it leaves the sample. Ιf we put a detector at the place where the first order' maximum is formed, we can observe the heating and cooling of the sample. Since we expect the cooling is exponential and that the time constant is inversely proportional to the thermal diffusivity, we can measure the thermal diffusivity of the sample. Figure 1 shows our experimental setup.

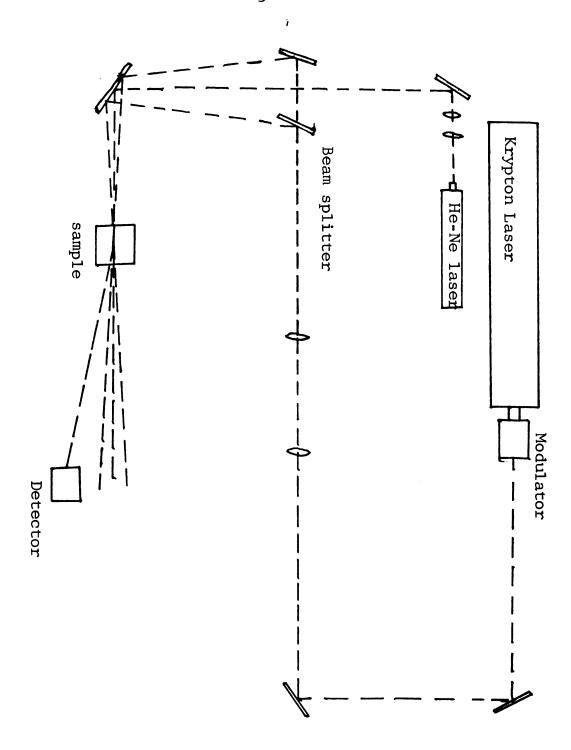


FIGURE 1 - EXPERIMENTAL SETUP

B. Lasers

In this experiment we are using two lasers. The first laser is Coherent Radiation - CR500 K Krypton Laser, used to heat the samples. The second is a small Helium-Neon laser, used as a probe. The Krypton laser has the ability to let the user choose from eight different wavelengths in the visible; two in the infrared and one in the ultraviolet. We are only interested in the visible range. In Table 1 are listed the wavelengths λ in nanometers, the color, and the maximum output power, in milliwatts. The ability to change the wavelength of the heating laser is important because the heating comes from the absorption of the laser light. Our samples have different absorption curves, therefore, a wavelength which is absorbed by one sample may be transmitted by another sample.

The second laser is a Spectra-Physics Model 120
Helium-Neon laser, which has a wavelength of 632.8
nanometers with a maximum output power of five milliwatts.
This laser light should be transmitted by the sample (should not be absorbed). This is the light which is diffracted by the grating created by the heating laser.

For the experiment to work the heating laser must be pulsed. This is done by a modulator. The one we use is a Coherent Associates Model 304 Acousto-Optic Modulation System. It is ideal for our use because it has the ability to put 75 to 80 percent of the laser power into the first order maximum.

TABLE 1 - POWER OUTPUT OF LASER

| Color | Wavelength Nanometers | Power Milliwatts | Measured Power* |
|-----------|--------------------------|---------------------|--------------------|
| Red - 1 | 676.4 | 120 | 25 |
| Red - 2 | 647.1 | 500 | 180 |
| Yellow | 568.2 | 150 | 100 |
| Green - 1 | 530.9 | 200 | 30 |
| Green - 2 | 520.8 | 70 | 90 |
| Blue - 1 | 482.5 | 30 | 8 |
| Blue - 2 | 476.2 | 50 | 30 |
| Blue - 3 | 468.9 | 5 | 5 |

*You only get the maximum power with all the laser mirrors clean.

The modulator is controlled by a waveform generator and two pulse generators. The waveform generator is used to generate a pulse. The rise and fall time is approximately one microsecond. It is used also to trigger one of the pulse generators. The waveform generator allows you to vary the repetition rate while the pulse generator allows you to vary the heating pulse width. The second pulse generator is used as a trigger for the computer and the oscilloscope. It also allows you the choice of triggering on the positive or negative edge of the heating pulse. The triggering pulse generator is triggered by the heating pulse generator. We are using Tektronix Type 162 Waveform Generator and Tektronix Type 163 Pulse Generator.

C. The Computer

We used a PDP 8/e computer to take and analyze data. The computer is equipped with an analog to digital converter which takes the analog signal from the detector, converts it into digital form and stores the numbers. The computer can take values which are between plus and minus one volts. The detector is nonlinear near zero volts, therefore we don't use negative values. This is where the oscilloscope comes in.

The oscilloscope is used to make sure that the signal from the detector is between zero and plus one volts. It is also used to adjust the equipment until we obtain the maximum signal size. The oscilloscope allows us to measure the size of the D.C. signal from scattered light from dirt on the cell walls, or small bubbles or large particles in the sample. By large particles we mean particles whose diameter is larger than five microns. This then allows us to adjust the D.C. offset on the detector to minimize the D.C. level. We can also use the oscilloscope to get an approximate value for \mathcal{T}_k , the time constant. From this we can tell which program to use in taking the data.

The computer has two basic programs for taking data. The first one is RM4 which has three versions. The difference between the three versions is the time between data points. See Table 2.

The RM4 series of programs was written by Mr. Daniel Edmonds.

TABLE 2 - RM4 PROGRAMS

| Program Name | Time Between Points In Microseconds | Data File |
|-----------------|-------------------------------------|--------------|
| RM4A5 | 3.6 | RM4EDA |
| RM4A6 | 7.2 | RM4G•DA |
| RM4A7 | 14.4 | RM4FDA |

The second data program is Basic Averager. Both programs do the same thing. The main difference is in the time between points. In basic averager we can choose any value between 30 microseconds and 4095 microseconds to be the time between points. Where in RM4 we have the choice of 3.6 microseconds, 7.2 microseconds, and 14.4 microseconds. The other difference is in the number of sums. In RM4 you can do 2^x number of sums. Where x is equal to any positive integer between 0 and 11. In Basic Averager you can take any integer number of sums up to 4095. This is because in RM4 you read in the number of sums from the switch register, while in Basic Averager the number of sums is read in from the teletype. Basic Averager came with the computer as part of the software.

The programs are designed to take the numerical value of each point every x microseconds. (x is the time you pick determined by the program you choose), and add this value to the sum of these points from previous scans. It repeats this process y times; where y is equal to the

number of sums you ask it to do. The program gives us the sum at each point. The reason we want to take the total sum is to eliminate noise. If you look at the signal from the detector on the oscilloscope you will see that it hops up and down. This hopping is caused by large particles inside the sample drifting through the beam. By taking a large number of sums, in some cases 2048, we can cancel out most of the noise, which gives us a nice smooth curve. Since the noise is random and the signal is always there, the randomness of the noise should cancel itself out.

For analyzing the data we have five interacting analyzing programs written in Fortran IV. The programs are basically the same. They differ only in the number of parameters and the parameters themselves used in fitting the data. The programs were written by Mr. Edward Grabowski. He took the program from a book. 1

The reason they are called interacting programs is because they allow you to interact with the computer. You can have the computer display the data, display the data plus the best curve superimposed, display their difference, and you can have it plot the data.

It is a least-squares fitting program which allows you to vary up to ten parameters. It is called Curfit: Program 11-5. The reason for five analyzing programs is that sometimes there are two times and if one time is much longer than the other then we can assume we had a single time

with a sloping base line. This is used when you are only interested in the shorter time. If you want the longer time or both times you want a program that will fit two different times. This is the purpose for the first three programs. One to fit a single time, one to fit a single time with a sloping base line, and one to fit two times. For the programs to work properly at least one third of the data must be base line.

The reason for the next two programs is caused by having the light being homodyne. In the program above we say that the light is heterodyne. By heterodyne we mean that the light causing the D.C. level of the detector has a definite phase with respect to the light causing the first order maximum. The detector is a square law device so that:

$$I_{het} \sim (E_{DC} + E_{signal})^{2}$$

$$since E_{signal} \sim E_{os} \exp(-t/\tau_{k})$$

$$I_{het} \sim E^{2}_{DC} + 2E_{DC} E_{os} \exp(-t/\tau_{k}) + E_{os}^{2} \exp(-2t/\tau_{k})$$

$$If E_{D} >>> E_{os} then:$$

$$I_{het} \approx E_{DC}^{2} + 2E_{DC} E_{os} \exp(-t/\tau_{k})$$

If there is no D.C. signal or if the D.C. signal has no definite phase relationship to the signal then we call the signal homodyne and using the same detector

$$I_{\text{homo}} \sim E_{\text{DC}}^2 \pm E^2 \text{sig}$$
 $I_{\text{homo}} \sim E_{\text{DC}}^2 + E_{\text{os}}^2 \exp(-2t/\tau_k)$

Thus we must be able to analyze data with two times \mathcal{T}_k and $\underline{\mathcal{T}_k}$ corresponding to a mixture of heterodyne and homodyne signals. This is the reason for the fourth and fifth programs. One to fit \mathcal{T}_k and $\underline{\mathcal{T}_k}$ with a straight base line and another to fit \mathcal{T}_k and $\underline{\mathcal{T}_k}$ with a sloping base line. For a listing of the programs see Table 3.

The reason we use these programs is because they can calculate τ_k much more accurately than we can by plotting the data points on semilog graph paper. Also they have a subroutine that calculates the difference between the data and its calculated curve. This is helpful because most times you can't see any difference by just looking at the fit. The computer displays the data and its calculated curve simultaneously superimposed on each other so you can see how good the fit is.

TABLE 3 - ANALYZING PROGRAMS

FIT TO PROGRAM NAMES A(1) + A(2) * EXP(-X/A(3))FAST 1 heterodyne; fitting one time A(1) + A(2) * EXP(-X/A(3)) + A(4) * XFAST 2 heterodyne; fitting one time with sloping base line A(1) + A(2) * EXP(-X/A(3)) + A(4) *FAST 3 EXP(-2X/A(3))heterodyne and homodyne; fitting τ_k and τ_k A(1) + A(2)*EXP(-X/A(3) + A(4)*EXP(-X/A(5))FAST 4 heterodyne; fitting two independent times A(1) + A(2) * EXP(-X/A(3)) + A(4) * EXP(-2X/A(3)) + A(5) * XFAST 5 heterodyne and homodyne; fitting au_{k} and $\frac{\boldsymbol{\tau}_k}{2}$ with a sloping base line

D. Detector

The detector is a silicon photodiode. The photodiode is a P-N junction diode. A diode can be thought
of as a resistor, which allows current to flow in one
direction. If you connect a battery to the diode so that
current tries to flow in the opposite direction, you
create a region around the P-N junction which is neutral.
Now if a photon enters this neutral region it can knock
an electron free from one of the atoms, forming an
electron-hole pair. The hole travels toward the negative
terminal of the battery and the electron travels toward
the positive terminal, causing a current.

The current is fed into a current-to-voltage converter which is an operational amplifier. From there it goes to a second operational amplifier which amplifies the signal. This operational amplifier is equipped with a D.C. offset. The amplifier is also designed to allow you to vary the gain. The photodiode is an EG & G SGD-040B. The electronics were designed by Mr. Edward Grabowski.

E. Furnace

One of my projects in this experiment was to build a furnace which would allow us to take data at temperatures other than room temperature. One of the design problems was that the furnace had to heat the sample and at the same time allow us to observe the sample. Also, our samples were rectangular in shape. The basic design of the furnace came from an article by G. G. Hacker, III, D. M. Eshelman, and R. L. Schmidt. Our furnace was a little simpler in design. Their furnace was designed to be able to control temperature fluctuations of the sample to plus or minus two millidegrees over a several hour period. We were only interested in maintaining a stable temperature with fluctuations of the order of plus or minus 0.25°C. The major difference between the two furnaces is that theirs was built for circular samples and our samples are rectangular. In Figure 2 there are some drawings of our furnace. When the furnace is connected to the controller we can obtain temperatures which range from room temperature to 300°C., theoretically. Because of the fact that we are using liquid samples we never go above 80°C. even though the furnace is designed to go up to 300°C. The controller we use was designed and built by Edward Grabowski. It has the ability to control the temperature to within 0.5° C in the on-off mode and theoretically it should control the temperature to plus or minus 0.1°C in the proportional mode. We only used the on-off mode which give the plus or minus 0.5°C needed in this experiment.

The controller in the on-off mode is basically a differential amplifier. When the reference voltage is less than the signal voltage the heater is on. When the reference voltage is equal to or greater than the reference voltage the heater is off. The only problem with this is that it takes some time before the controller can tell that the furnace is being heated. Therefore, there is overshooting and undershooting of the set temperature. This causes the temperature to oscillate about the desired temperature.

In the proportional mode, the furnace is heated in pulses, not continuously as in the on-off mode, where the heating pulse becomes shorter as the furnace reaches the set temperature. This is done by sending in a ramp signal. To have the mode working correctly you must know the time constant of your furnace. By time constant we mean the time interval between the time you put heat into the furnace and the time the controller sees a change in temperature. The furnace uses a thermistor to measure the temperature.

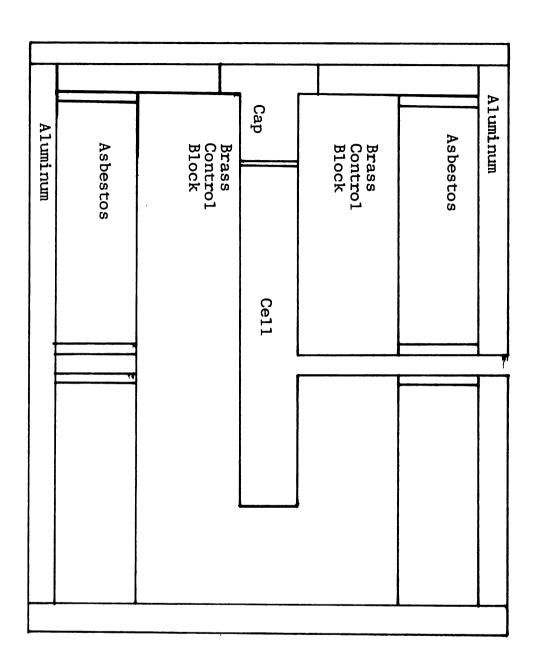


FIGURE -2A- CROSS-SECTIONAL VIEW OF FURNACE

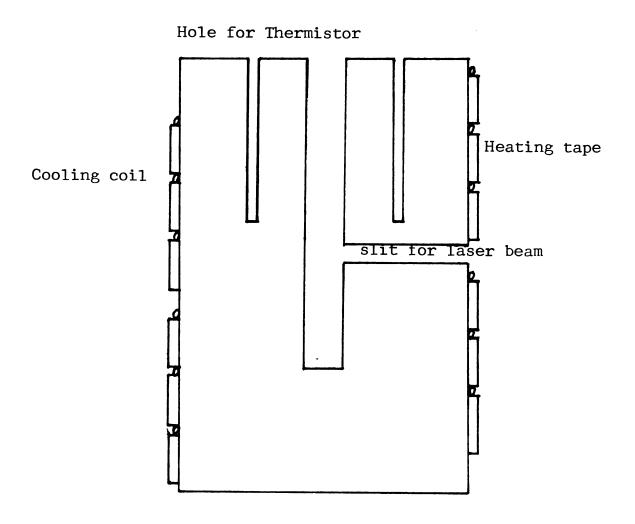


FIGURE -2B- CROSS-SECTIONAL VIEW OF CONTROL BLOCK

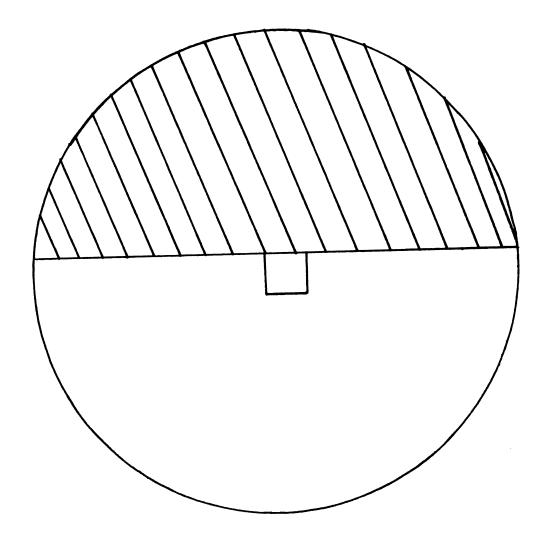


FIGURE -2C- TOP VIEW OF BRASS CONTROL BLOCK AT BEAM LEVEL

F. Measurement of d

On our beam splitter is a micrometer. The micrometer is used to vary the distance between the mirrors. We use the readings on the micrometer to determine d (spacing between fringes). Following is the theory that justifies this.

The way we obtained a grating is by splitting the heating beam into two equal beams and letting them intersect inside the sample. This will cause a diffraction pattern in the region where they overlap. The spacing between the fringes, d, is equal to $d = \frac{\lambda}{\sin(\theta/2)}$ for small angles $\sin(\theta/2) \approx \theta/2$. See Figure 3. Therefore $d = \frac{\lambda}{\theta}$ where θ is defined as the angle between the two beams. But for small angles $\theta = x/D$ where x is defined as the distance between the two mirrors of the beam splitter, and D is the distance between the beam splitter and where the two beams intersect. This leads to the equation:

$$d = \frac{\lambda D}{x}$$

If we let x = Y - C, where C is equal to a constant and Y is equal to a micrometer reading which is attach to the beam splitter. C is put in the equation, because the micrometer doesn't read zero when the distance between the mirrors is zero. Therefore $d = \frac{\lambda D}{Y - C}$

We can measure D and we select λ . Therefore if we measure d and plot $\frac{1}{d}$ using our least square program we can get C and Y from the program. Now if we plot d vs. $\frac{\lambda D}{Y-C}$

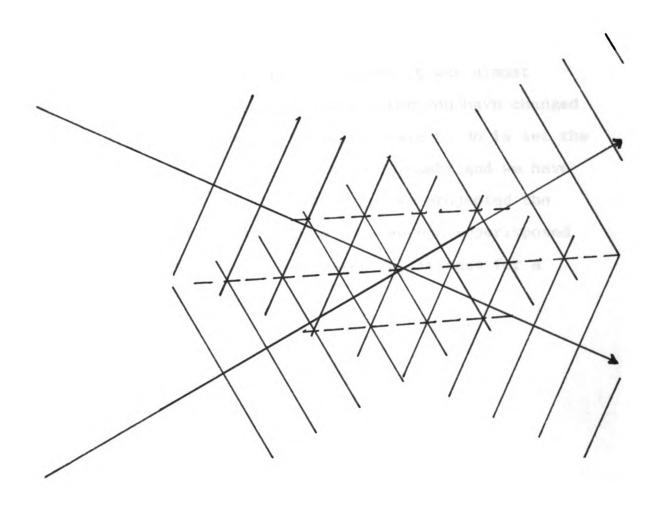


FIGURE 3 - GEOMETRY OF THE TWO INTERSECTING BEAMS

--- Is the fringes

we get a curve that translates the dial setting on the micrometer into d (fringe spacings).

This has a great advantage over the old system of measuring fringes. In the old system it was almost impossible to get the same d back after you have changed it. But with the new system all we have to do is set the micrometer to the setting it was previously and we have the same d value. In the old system we projected the image of the intersecting beam on a screen superimposed over a scale and counted the number of fringes for a certain length.

G. Theory

The thermal diffusivity of a sample which has one dimensional heat flow can be derived as follows.⁴ The total amount of heat entering the differential face dxdz at y is given by dQ = -dxdz $(K(\frac{dT}{dy}))^{dt}$ See Figure 4.

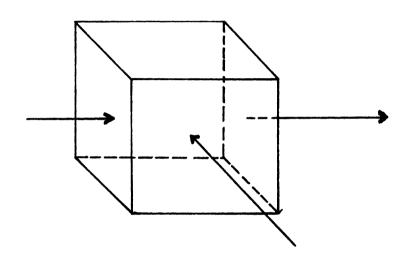


FIGURE 4 - VOLUME ELEMENT dxdydz

where K is defined as the thermal conductivity and Q is defined as heat. To find the amount of heat leaving the element at y + dy, let

$$F(y,T) = K(\frac{dT}{dy})$$

By Taylor's series expansion:

$$F(y + dy,T) = F(y,T) + \frac{dF}{dy} dy$$
$$= K \frac{dT}{dy} + \frac{d}{dy} (K \frac{dT}{dy}) dy$$

This leads to the fact that

$$dQ (y + dy) = dxdz (K \frac{dT}{dy} + \frac{d}{dy} (K \frac{dT}{dy}) dy) dt$$

If we add to this dE, which is defined as the total quantity of heat which goes into increasing the internal energy of the volume element:

$$dE = CP dxdydz \frac{dT}{dt} dt$$

where C is defined as the specific heat, and P is defined as the density. Also if we add dQ_g which is defined as the total amount of heat generated in the volume element. Where,

$$dQ_g = q$$
" $Vdt = q$ " $dxdydzdt$

where q" is the rate at which heat is being generated internally. Therefore using conservation of energy we have:

$$dQ(y) + dQ_g - dQ(y + dy) - dE = 0$$

 $dQ(y) + dQ_g = dQ(y + dy) + dE$

This leads to

-dxdz (K
$$\frac{dT}{dy}$$
) dt + q" dxdydz dt
=-dxdz ((K $\frac{dT}{dy}$) dt + $\frac{d}{dy}$ (K $\frac{dT}{dy}$) dy) dt + CP dxdydz $\frac{dT}{dt}$ dt

Divide by dxdydzdt which reduces to

$$q'' = -\frac{d}{dy} (K \frac{dT}{dy}) + CP \frac{dT}{dt}$$

Therefore,

$$q'' = -K \frac{d^2T}{dy^2} + CP \frac{dT}{dt}$$

$$q'' + K \frac{d^2T}{dy^2} - CP \frac{dT}{dt} = 0$$

which leads to

$$\frac{d^2T}{dy^2} + q''/K = \frac{CP}{K} \frac{dT}{dt}$$

where $D_{Th} = \frac{K}{CP}$ which is defined as the thermal diffusivity.

Therefore:

$$\frac{d^2T}{dv^2} + \frac{1}{K} q'' = \frac{1}{D_{Th}} \frac{dT}{dt}$$

where in our case $q'' = \theta I_e$, where θ is defined as the light absorption coefficient of the sample. I_e is defined as the incident intensity. Therefore;

$$\frac{1}{D_{Th}} \frac{dT}{dt} - \frac{d^2T}{dv^2} = \frac{1}{K} \Theta I_e \quad \text{(equation 1)}$$

If the depth of penetration (θ^{-1}) is very large compared to d (the distance between the fringes), and if d is very small compared to the sample thickness, then at the end of the heating pulse (t=0) there is a temperature distribution which varies with position.

$$\Delta T (0,y) = \overline{T} (0)\cos ky$$
 (equation 2)

T is the temperature amplitude and $k = \frac{2\pi}{d}$.

Now if we assume a solution to equation 1 for time t=0, that is $I_e=0$, then if we replace T with ΔT using equation 2 then:

 $T(t,y) = T(t) \cos ky = T(0) \exp(-t/\tau_k) \cos ky$

Now if we insert this into equation 2, we have:

$$\frac{d^2}{dy^2} \left(\Delta T \right) - \frac{1}{D_{Th}} \frac{d(\Delta T)}{dt} = 0$$

Therefore:

$$\frac{d^2}{dy^2} \quad (\Delta T) = \frac{1}{D_{Th}} \frac{d(\Delta T)}{dt}$$

$$\frac{d^2}{dy^2} (\overline{T} (0) \exp(-t/\tau_k) \cos ky)$$

=
$$\frac{1}{D_{Th}} \frac{d}{dt}$$
 (\overline{T} (0) exp(-t/ τ_k) cos ky)

Now take the derivative of both sides

$$-k^{2} T (\theta) \exp(-t/T_{k}) \cos ky$$

$$= \frac{-1}{D_{Th}T_{k}} \overline{T} (0) \exp(-t/T_{k}) \cos ky$$

which reduces to

$$-k^2 = \frac{-1}{D_{\text{Th}} \tau_k}$$

which leads to $\tau_k = \frac{1}{D_{Th} k^2}$

but
$$D_{Th} = \frac{K'}{PC}$$
 and $k = \frac{2 \sqrt{r}}{d}$

therefore:

$$\tau_k = \frac{PC}{K} \left(\frac{d}{2\pi}\right)^2$$
.

The temperature decreases exponentially with a time constant of \mathcal{T}_k , which depends on d^2 , and $1/D_{Th}$ or d^2 , P, C, and 1/K. This shows that if we measure the relaxation time of a liquid, and know what d is, we can calculate the thermal diffusivity of the liquid. And from the thermal diffusivity we can obtain the thermal conductivity of the liquid.

When the two beams meet there is a temperature grating formed. Since the liquid has a temperature variance which can be approximated as

$$T = \overline{T} \cos ky$$

We can say it has an index of refraction that varies as $\Delta n = -n^{2}$ (1 + cos ky)

where n' is the peak value for the change in the index of refraction divided by two.

This is caused by the fact that when the sample is heated its density changes which changes its index of refraction.

The next question is why this change in density doesn't change \mathcal{T}_k since \mathbf{D}_{Th} (the thermal diffusivity) is dependent on density. This is because heating the sample we only change the temperature a few millidegrees. Therefore, the change in the density is very small.

If we have a plane wave in the region of the heat grating we will see that some of the light is refracted. See Figure 5.

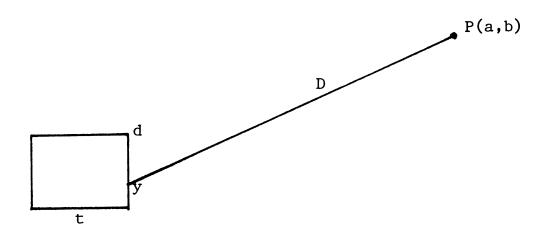


FIGURE 5 - DIFFRACTION GEOMETRY

The light intensity at a point P(a,b) outside of the sample can be determined as follows:

$$D = \sqrt{a^2 + (b-y)}$$

Where D is the distance from the heat grating to point P(a,b).

a is the x component of D and b is the y component of D measured from y = 0.

Let $n = n(y) = n_0 - n! (1 + \cos ky)$.

The phase shift through the medium:

$$\Theta (Y) = \frac{2\pi}{\lambda} t n(Y)$$

where t = the thickness of the temperature grating. λ is the wavelength of the transmitted beam.

The total phase shift of point P is;

$$\theta \text{ total} = \frac{2\pi t}{\lambda} \text{ n(y)} + \frac{2\pi}{\lambda} \sqrt{a^2 + (b-y)^2}$$

at $\Theta = 0$ plane, let $\overline{E} = E_0$

Also lets assume D>> d.

Therefore at point P;

$$I = \left| \int_0^d \exp (i \theta \cot a) dy \right|^2$$

I $(a,b) = \left| \int_0^d \exp\left(i \, \frac{2 \, \pi - t}{\lambda} \, n(y) + \frac{i \, 2 \, \pi}{\lambda} \, \sqrt{a^2 + (b - y)^2} \right) \, dy \, \right|^2$ There is no closed form solution to this equation but you can get a numerical solution. All we are interested in is the relative position of point P and we can get a good approximation of this by assuming that in the sinewave is a step function. If we let the sinewave be a step function and assume that the grating is caused by intensity variation and not phase variations our problem reduces to the one of a classical multislit diffraction grating. The major difference is when you take the Fourier transform of a sinewave you get a sinewave back, but the Fourier transform of a step function is a family of sinewaves. Therefore by replacing a sine function with a

step function the theory will predict a first order maximum,

a second order maximum, a third order maximum, and so on.

In our case all we should get is a first order maximum.

The reason we feel that the grating is caused by an intensity variation and not a phase variation is because if it were a phase variation a change in the intensity of the heating beam should change the position of the first order maximum. We never saw this change in position.

III. RESULTS

There were two things which we tried to do in this experiment.

- Demonstrate that the apparatus and furnace worked and estimate the accuracy of measurement.
- 2) Determine if we could see the effect of rather large amounts (up to 5%) of a high molecular weight polymer on the thermal diffusivity of water-polymer solution.

The apparatus including the furnace did work, it was possible to take data for d ranging from 15 microns to 120 microns, and at temperatures from room temperature to 80° C. If the samples were clean and if not much local heating of the samples occur we can get consistent results to within 1% for fixed d. If we vary d, the results are consistent to several percent which is the accuracy to which we can measure d. Table 4 gives a set of results obtained on pure water, 1%, 2.5%, and 5% polymer at fixed d = 50.3 microns and T = 22° C. Data was taken with RM4A6 and analyzed at FAST 2. The variation is less than 1%. This demonstrates that we did not observe any effect of the polymer on the thermal diffusivity of the solution in this concentration range.

If we compute the thermal diffusivity of the solution:

$$D_{\text{Th}} = \frac{1}{4\pi^2} \frac{d^2}{\tau_k} = \frac{1}{4\pi^2} \frac{(50.3 \times 10^{-4} \text{cm})^2}{374 \times 10^{-6} \text{ sec}}$$

$$= 1.71 \times 10^{-3} \text{ cm}^2/\text{sec}$$

Compare it with

 $D_{\overline{Th}} = \frac{K}{PC}$ obtained from data on water in the Handbook of Chemistry and Physics.

$$D_{\text{Th}} = \frac{1.43 \times 10^{-3} \text{ cal/sec} - \text{cm} - {}^{\text{O}}\text{K}}{(.997 \text{ gm/cm}^2) (.998 \text{ cal/gm} - {}^{\text{O}}\text{K})}$$
$$= 1.44 \times 10^{-3} \text{ cm}^2/\text{sec}$$

We see that the difference is rather large, approximately 19 percent. We do not understand this large discrepancy but it may come in part from air dissolved in the water and in part from a possible error in d.

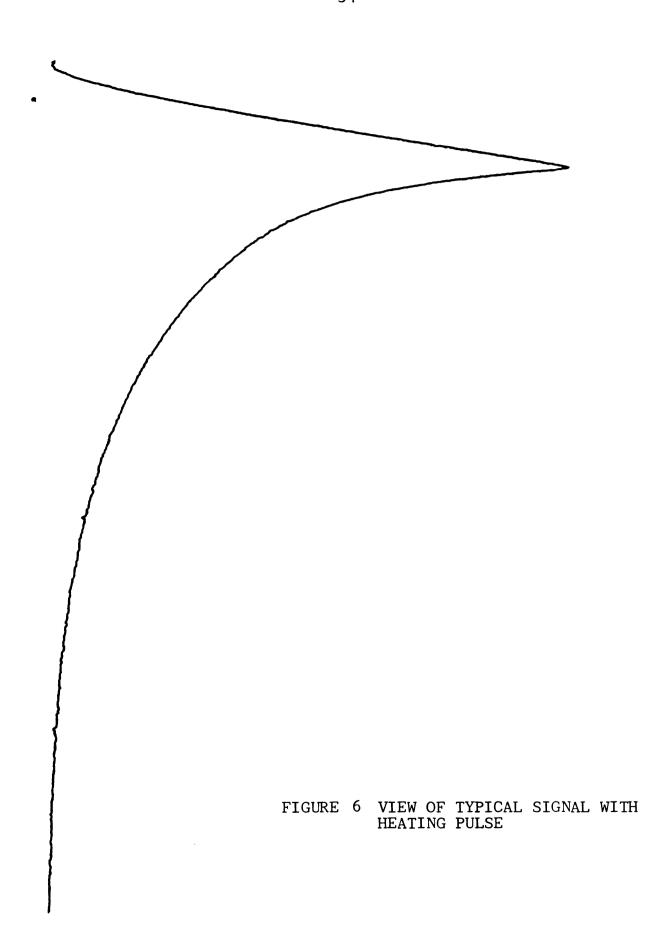
The solution of polymer in water with a little methylred to absorb the light gave interesting results which we now think we understand. We saw two relaxation times, au_{s} (\sim 400 microseconds) and au_1 (many milliseconds). $au_{f s}$ was dependent on d², was independent of temperature or of the polymer concentration. au_1 was independent of d, strongly dependent on temperature and on polymer concentration. After many experiments with different concentrations of polymer and also with the dye dissolved in water whose pH we changed by adding an acid or a base, we found that the long time was apparently due to the bleaching of the dye. au_1 was very long in basic solutions and vanished in acid solutions. (The color of the solution also changed). We think that au_1 was the time for recovery of the dye and therefore it did not depend on d. apparent dependence of au_1 on polymer concentration was

due to the fact that the polymer was strongly basic. A 1% solution has a pH of 10.3 so that changing the concentration also changed the pH. We do not understand why $alpha_1$ depends so strongly on either pH or temperature but it must be related to the change of optical and chemical properties of this organic dye.

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TABLE 4 - DATA AT 22°C

| % Polymer | <u>Microseconds</u> |
|-----------|---------------------|
| 0 | 376 |
| 1% | 375 |
| 2.5% | 376 |
| 5% | 371 |
| Average | 374 <u>+</u> 2 |



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