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Final Report

Thermally Induced Interference Patterns in Liquids

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I. Introduction

In the fall of 1967, the Sandia Corporation awarded to Seymour S. Alpert a contract (#53-6574) entitled "An Investigation into the Cause of Self-Induced Divergence of Laser Light in Liquids." During the course of this previous contract, it was determined that the divergence of light in liquids resulted from the setting up of thermal effects in these liquids. A further contract (#53-8386) was awarded by the Sandia Corporation in order to determine the nature and consequences of the light induced thermal effects in liquids. The title of this latter contract is appropriately "Thermally Induced Interference Patterns in Liquids."

Many observations were made of thermal-light phenomena. The most striking of these phenomena was the "blooming" of the original laser light beam into a complex ringed structure on passing through an energy absorbing liquid (Fig. 1). In order to make quantitatively reliable measurements of the size and angular divergence of laser light going into the "bloom" it was necessary to devote a major output of time and energy into the development of techniques designed to obviate problems of noise and instability relating to the optical interference pattern.

These developmental procedures are described in this report as are the general over-all results of the year's work. The controlling of the various factors that cause optical

pattern instability has been so successful as to cause comment from visitors as to the marked stability of the optical pattern. It should be kept in mind that this stability is not ordinarily expected in a phenomenon occurring in a liquid where thermal convection and microphonic currents can cause large scale changes in the observed results.

II. Method

The experimental arrangement was extremely simple. (See Fig. 2.) The laser beam was focused by a spherical dielectrically-coated mirror into the energy absorbing liquid contained in a thin cell. The resulting optical pattern then fell on a CdS photodetector which was mounted on a lathe bed so that the pattern or parts of the pattern could be traced. The detector was behind several variable light attenuators the function of which was to keep the light intensity which reached the detector at a sufficiently low level so that the detector response was linear in regard to intensity. The half-wave plate shown in Fig. 2 was used solely as an intensity controlling device since it varied the plane of the laser polarization that fell onto the metal-coated flat deflecting mirror. The use of the half-wave plate allowed for a 5% change in the laser energy that reached the cell.

The cell itself was either a 4° or 10° aluminum wedge with quartz sides. The wedge shape allowed for the thicknesses

of liquid to be changed continuously. The concentration of the absorbing dye could be changed; the resulting liquid was introduced into the quartz cell and a study made of the optical pattern size as both a function of fractional light absorption and cell thickness.

III. Developmental Techniques

As was mentioned previously, it was necessary to develop techniques to obviate problems of nonreproducibility and non-stability. The major developments to improve accuracy are listed as follows:

A. Temperature stability

The initial thermally induced interference patterns had been observed originally by "doping" various organic liquids with I_2 . A metallic cell enclosure having a large heat capacity was constructed. The I_2 -doped liquid was placed in a cell in the enclosure. The size of the pattern was studied as the enclosure temperature was varied. There was a marked dependence of pattern size on the ambient temperature. This effect was felt to result from a decrease in the population of absorbing iodine molecules as the temperature increased; it is believed that the iodine molecules differentially diffused out of the heated region. Various dyes were investigated in order to find one which would be nearly temperature independent. A solution of malachite green in acetone was found to be quite satisfactory.

B. Filtering

The heating effects caused by laser light absorption are known to cause convection currents. In one of the geometrical arrangements which we use light enters the cell from below and convection currents are present; these currents trap small dust particles at the point of the entry of the light into the cell. Such trapping decreases in an unknown fashion the amount of light absorption in the cell. Dust particles were found to be eliminated satisfactorily by triple filtering using medical syringe filters and fine filter paper. When light entered the cell from above, dust trapping was not evident.

C. Mirror Focusing

In order to focus the laser light into the cell containing the energy absorbing liquid, a 50 cm concave dielectric mirror was used; however, it was found that maximum effects occurred when the liquid samples were placed at 57 cm from the reflecting mirror. This is readily understood in view of the small laser beam diameter and the small (0.2 mrad) angular divergence of the laser beam. When the thermally induced optical pattern size was studied as a function of the cell-to-mirror distance it was found to peak at 57 cm with a broad maximum. This being the case, cells are now placed at 57 ± 1 cm from the mirror.

D. Laser Stability

It was discovered that mirror tuning of the laser to

give a constant power output did not always result in the same size interference patterns. Investigation led to the conclusion that although the output power was held constant, the mode structure was slightly different for each different mirror configuration. Different mode structure would have different intensity densities which when focused down into the light-absorbing liquid cell would allow for a varying temperature distribution.

The solution to this problem was attempted by the following steps: (a) optimization of the laser mode structure, (b) locking the mirrors at their optimal positions, and (c) then waiting $1\frac{1}{2}$ hours for each warm up period to allow the mirrors to assume their final positions.

E. Horizontal Cell Orientation

Since convection currents are present and since these currents modify the optical pattern, it is important that the normal to the cell be perpendicular to the horizontal gravitational plane. A non-horizontal condition allows for a vertical component of flow to exist. Achieving a horizontal configuration is accomplished by using a plumb line to orient the incoming light beam and then to bring the first surface reflection back on itself.

This process assures that the convection currents present equally affect all parts of the optical pattern.

Experimentation bears out the improved reliability of present measurements where the preceding precautions have been taken.

F. Power Measurements

Since all the optical pattern data is taken at a standard, fixed input power setting, it is important that the standard input power be measured reliably. In the past this was done making use of a solar cell as a detector and placing the solar cell at the input region of the tested liquid cell where the laser light is concentrated into a very tiny bright spot of about 0.02 cm diameter. Unfortunately two effects open this procedure to doubt; namely, there is detector saturation caused by the high light intensities and also there is a nonuniformity of detector response in tiny regions of the size of the bright laser spot. To improve the detector response, a guide was constructed and placed in a position such that the laser beam which would have entered the cell would have diffracted sufficiently so as to cover an area of about 1/10 that of the solar cell detector. This averages out local variations and avoids any saturation problems.

IV. Results

A wide variety of interesting results have been obtained. Figures 3 and 4 show the dependence of the steady state maximal angular radius of the interference pattern on the cell thickness and on the fractional absorption, respectively. The dependence of the pattern size on cell thickness is not extremely marked

subject to the condition that the fractional absorption be constant. This point will be discussed further in the following section dealing with theoretical considerations. It is to be noted that, however, for cell thicknesses less than 1 mm and for fraction absorptions of greater than 80% the pattern size falls off very quickly for small thickness. For heavy "dopings," the pattern size falls off very gradually for increasing thickness.

The previous results are shown for an arrangement of the apparatus wherein the laser light enters from below. It had been noticed that the behavior of the interference pattern changed if the geometry was altered so that the entering laser light came from above. A typical curve showing this difference in behavior is shown in Fig. 4. The up-down asymmetry is believed to be caused by convection currents which, in the case of light entering from above, tend to be less than the case where light enters from below. In the latter case convection currents are caused by virtue of a warm less-dense liquid forming below and rising.

A modification of the experimental arrangement of Fig. 2 was adopted to study in a controlled manner the effects of liquid velocity on the optical pattern and structure. The cell under study had a rectangular cross section 1 mm thick and 1 cm wide. The "doped" liquid (malachite green in acetone) was allowed to enter and exit from two-end openings in the

cell. The angular diameter of the resulting optical pattern is shown in Fig. 5 as a function of liquid velocity. In addition to the results presented in Fig. 5, a flattening of the "upstream" side was noted. This type of behavior had been noticed in the case where a thin cell had been exposed to light with the cell plane vertical. In this previous case, the introduction of lint which was illuminated with side-light allowed a flow pattern to be determined visually. In the case at hand, the flow lines are presumed to be reversed from that of the vertical plane and are shown schematically in Fig. 6. It is a premise of the theoretical approach to the general problem of thermal lens production in liquids that liquid motion negates a stationary thermal distribution necessary for a lens effect. The faster and closer together are the flow lines, the less will be the lens effect.

Further qualitative proof of the negative effects of convection comes from the fact that viscous liquids such as cyclohexane and canada balsam when doped do not show the deleterious effects of liquid motion resulting from different geometries. Their resistance to motion allows for a more stable pattern configuration.

V. Theoretical Considerations

In addition to the experimental points previously listed, some successful theoretical work has been accomplished. The wedge data obtained has shown that the size of the thermally

induced optical interference pattern is dependent on the thickness of the cell all other parameters being held constant. A sharp break occurs in this behavior for cells the thickness of which are greater than 1 mm. For these thicknesses the laser spot size ω_0 is considerably smaller than the cell length; in this case, the heat equation can be solved cylindrically and the laws of geometric optics used so as to predict the optical angle of refraction for any laser ray passing through the liquid cell as a function of its radial position. The functional relation is given by

$$\theta = \frac{pP}{2\pi} \frac{1}{n} \frac{1}{k} \frac{dn}{dT} \left[1 - \exp - \left\{ \frac{2r^2}{\omega_0^2} \right\} \right] \dots \quad (1)$$

where θ is the angle of refraction, p is the fractional power absorption, P is the total laser power in cal/sec, n is the index of refraction, k the thermal conductivity, dn/dT is the thermal index gradient, r the radial position of a ray, and ω_0 is the laser beam spot size. Equation (1) can be maximized to find the extreme semi-radial angle for an optical pattern regardless of its structure. Optimization yields

$$\theta_{\max} = \frac{.902}{2\pi} \frac{pP}{n} \frac{1}{k} \frac{dn}{dT} \frac{1}{\omega_0} \dots \quad (2)$$

The validity of Eq. (2) has been checked numerically against the experimental data and works remarkably well. All the

various dependencies shown in Eq. (2) have been checked and seem to be valid. The spot size, ω_0 , appears in the denominator. This is essentially a statement of power density. Presumably a more powerful laser focused into a smaller spot will show appreciably greater effects. Eq. (1) is similar to a relation derived by J. P. Gordon et al.¹ for the spatial temperature dependence.

The validity of Eq. (1) breaks down for very thin (less than .5 mm) or very thick (greater than 3 mm) cells. In the former case the boundary conditions are different than those used in the derivation of Eq. (1). In the latter case, the conical light distribution lowers the energy density to the point where the effect begins to fall off.

A calculational approach has been initiated to improve on our theoretical understanding of the interference effect. This present theoretical approach is not yet completed; hence, this must be a preliminary report. The validity of both Eqs. (1) and (2) seems assured for thin cells which have cylindrical symmetry and for which the end surfaces are far removed from each other in comparison to the laser spot size, ω_0 . This latter condition is removed if the energy cylinder in the liquid has end surfaces which are separated by a differential distance and if the temperature change from one differential section to another is small. The liquid is divided into a set of lamella. The incident energy

¹ J. P. Gordon, R. C. Leite, R. S. Moore, S. P. S. Porto, and J. R. Whinnery, J. Appl. Phys. 36, 3 (1965).

is known to have a Gaussian distribution. The Gaussian distribution of the first lamellar section is divided into many (say 100) equal energy sections. This division defines an equal number of light rays which mark the boundary of each energy division. These light rays are traced through the first lamellar section. Since each light ray is associated with a fixed and equal amount of energy, the energy distribution is known in the second lamellar section. If the energy density is known in the second lamellar section, a modified Newton's heat equation

$$dT/dr = \frac{1}{2\pi\ell k} \frac{1}{r} \iint_0^{\ell r} Q(r',z) dr' \dots \quad (3)$$

can be solved to give an equation analogous to Eq. (2) but with an other than Gaussian source function $Q(r',z)$. In Eq. (3), ℓ represents the cell thickness and z the thickness variable. $Q(r',z)$ has units of energy per time. This process is then repeated for the next lamellar section and so forth.

The main problem must be an iterative one because the heat distribution and light refraction are not independent, each affecting the other.

The computer code which is generally described in the previous paragraphs, has been written and does show the general behavior demonstrated by the optical pattern. It does not show, however, the location of the interference fringes. This fault will be corrected by introducing into the program currently being worked upon a factor which will allow us to keep track of the relative phase changes between different rays which come

together in the far field. Whether or not this will show the complete light extinction observed at the pattern nulls, remains to be seen.

VI. Miscellany

The following interesting facts are offered with no explanations at the present time.

A. Popping

For cell thicknesses less than .2 mm and for "dopings" greater than 80%, cavitation occurred with a resounding violent audible shock. This situation was avoided since some concern was felt for the state of the cell. It was felt that the "popping" was so violent it might cause damage to the cell.

B. Bubble pulling

In preparing samples of acetone doped with malachite green, small bubbles of air about .5 mm in diameter were often formed. If these tiny bubbles were illuminated directly with the focused laser beam, they were trapped and could be moved in any translational manner by moving the laser beam.

Captions

Figure 1. Steady state optical pattern caused by vertical focusing from below of 62 mW of 6328 Å laser light into a 2 mm quartz cell containing CS₂ and I₂. Fractional absorption of .63. The maximal angular diameter is about 210 mrad.

Figure 2. The experimental arrangement.

Figure 3. The maximal angular radius vs cell thickness.

Figure 4. The maximal angular radius vs the fractional absorption.

Figure 5. The optical pattern size vs the velocity of liquid flow.

Figure 6. Presumed flow lines of liquid in region of laser light. Laser light region is indicated by circle.

Fig. 1

Figure 2

10 Millimeters to the Centimeter

Figure 3

FIG 4

MALACHITE GRN
IN ACETONE

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1mm THICK

FRACTIONAL ABSORPTION
OF .54

FIGURE 5

FIGURE 6