DISPERSION OF THE NONLINEAR REFRACTIVE INDEX OF CS$_2$

IN THE SPECTRAL RANGE OF 9-11 µm

DISSERTATION

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The nonlinear refractive index ($n_2$) of room temperature liquid CS$_2$ in the wavelength range of 9 to 11 micrometers is measured. A line tunable hybrid CO$_2$ TEA laser and amplifier system is used for the experiments. In these measurements the well known photoacoustic method is utilized to observe the onset of whole beam self-focusing. The photoacoustic signal in a CS$_2$ cell, much longer than the confocal parameter, is monitored. The departure of the acoustic signal from linear growth marks the critical power for the onset of nonlinearity. It is experimentally verified that the phenomenon is power dependent as expected from self-focusing theory.

The value of $n_2$ is then calculated from the theoretical model of self focusing. Measurements of the on-axis irradiance transmitted through the nonlinear material as well as the measurements of beam distortion are used to verify the validity of the photoacoustic method. In all the measurements the on-axis intensity was smaller than the calculated threshold intensity for stimulated Brillouin scattering. The back reflection was monitored to make sure that stimulated Brillouin scattering was not playing a role in the phenomenon.
In our preliminary study, the value of $n_2$ in CS$_2$ at 10.6 $\mu$m was measured to be an order of magnitude larger than the reorientational $n_2$ in the visible and near infrared. Further study of the nonlinear refraction in CS$_2$ at different CO$_2$ laser lines shows that $n_2$ varies by a factor of four with its maximum of $(11.6 \pm 2) \times 10^{-10}$ at 10.24 $\mu$m. Measurements of $n_2$ at different temperatures and different polarizations support calculations which show electrostriction as the dominant microscopic phenomenon responsible for the nonlinearity. At the same time the polarization dependence of the nonlinearity hints to the presence of a small reorientational Kerr effect. Other experimental and theoretical evidence rules out the presence of significant thermal defocusing in the measurements. The possible origin of the observed dispersion is discussed and future experiments are suggested.
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CHAPTER I

INTRODUCTION

The nonlinear refractive index ($n_2$) of room temperature liquid CS$_2$ in the wavelength range of 9 to 11 micrometers is measured. While nonlinear refraction in CS$_2$ has been studied$^{3-7}$ extensively in the visible and near infrared spectral region, there have been only two$^{8-9}$ reported measurements of $n_2$ in CS$_2$ at 10.6 $\mu$m. Reasonable agreement is found among the various measurements of $n_2$ in the visible and near infrared; however, the values reported at 10.6 $\mu$m differ by three orders of magnitude. The study of nonlinear refractive properties of CS$_2$ in the visible and near infrared has shown that there is no dispersion in this spectral range. Such systematic studies in the spectral range of 9 to 11 $\mu$m have not been attempted before. In view of the importance of CS$_2$ as a standard against which nonlinear materials can be characterized in the infrared, and in order to resolve the existing discrepancy in the literature, the present study has been undertaken. There is a host of different techniques for the study of the nonlinear refraction. Pump-probe type of experiments with their inherent complications both in performing the experiment and interpreting the results$^9$ will be briefly discussed in Chapter IV. Interferometric$^{10}$ and
ellipse rotation⁴,¹¹ methods have been utilized in the visible spectrum to measure the nonlinear refractive index of a variety of liquids and solids. Many different methods have been used to detect the onset of self-focusing and self-defocusing, thereby to measure \( n_2 \). Among these are measurements of intensity on the axis,⁶,⁷ and the beam shape transmitted through the nonlinear material.⁶⁻⁸ Also measurement of the power threshold for the stimulated Raman emission has been used.¹²,¹³ However, in the present study three different techniques were used. We will limit our discussion to these three methods, to which we will refer as the photoacoustic, power limiting and beam distortion measurement methods. Complete descriptions of these techniques are given in Chapter III. In the photoacoustic method, the onset of whole beam self-focusing is detected by monitoring the photoacoustic signal in the sample using a piezoelectric transducer coupled to the wall of the \( \text{CS}_2 \) cell. The departure of the acoustic signal from linear growth marks the critical power for self-focusing. In this method it is assumed that the sharp rise in the acoustic signal corresponds to the critical power for self-focusing. The abrupt increase in the photoacoustic signal can in principle be caused by other mechanisms such as material breakdown or stimulated Brillouin scattering. The validity of this assumption was experimentally verified. It will be shown in
Chapter III that the material breakdown is a consequence of self-focusing and that stimulated Brillouin scattering is not playing a role in these measurements.

As suggested in the above paragraph, self-focusing can be used to measure the nonlinear refractive index of materials that exhibit this nonlinearity. In a nonlinear material with cubic nonlinearity such as CS$_2$ index of refraction can be written, in steady state approximation, as

$$n = n_0 + \frac{1}{2} n_2 |E(r,t)|^2,$$

where $n_0$ is the unperturbed refractive index, $E$ is the electric field amplitude, and $n_2$ for an isotropic material is related to the third order nonlinear susceptibility by

$$n_2 = \frac{(12\pi/n_0)}{X_3(\omega;\omega,\omega,-\omega)}.$$

The factor 12 in definition of $n_2$ includes a degeneracy factor of 3 which arises because there are three identical terms in which $-\omega$ occupies one of the last three arguments. With a nonuniform beam profile, the change of index of refraction induced by the beam maps the nonuniformities of the beam itself. The induced index profile in turn affects the linear propagation of the beam. If $n_2$ is negative, the induced index profile will act as a negative lens leading to self-defocusing. If $n_2$ is positive, it leads to self-focusing, which ends with catastrophic collapse of the beam to a mathematical singularity. What occurs beyond singularity is of no practical significance because even before reaching
the singularity, the intensity in the beam is large enough that either material breakdown or higher order nonlinearities enter the picture. The simple cubic model will "break down." This process has a sharp threshold in power. Marburger has solved the nonlinear wave equation\(^2\) and gives the critical power for self focusing of a Gaussian beam to be

\[ P_2 = 3.77 \frac{c\lambda^2}{32 \pi^2} n_2 \]

where \( c \) is the speed of light in the vacuum, and 3.77 comes from numerical computations. It is this formula that connects the measured critical power for self-focusing to the reported values of \( n_2 \) in this work. In arriving at this relation it was assumed that the response of the material is instantaneous. The validity of this relation has been verified experimentally by previous workers.\(^6\) Change of index of refraction due to thermal induced density fluctuation has a slow response, because it is related to the thermal expansion and thermal conductivity. The former has a response time of the order of hundreds of microseconds, and the latter has a response time of the order of a few milliseconds. For our pulses these effects are too slow to be significant. The reorientational nonlinearity in \( \text{CS}_2 \) has a fast response time of 2 ps.\(^14\) The response time of the electrostriction effect on the other hand depends on the beam size, speed of sound, viscosity and material compressibility. For \( \text{CS}_2 \) and our experimental conditions this response is
approximately 30 ns. Under these conditions and for 270 ns long pulses used in our experiment, the steady state assumption (instantaneous response) is not unreasonable.

The magnitude and sign of \( n_2 \) is determined by the material properties and the fundamental microscopic processes responsible for the nonlinearity. The electrostriction and reorientational Kerr effect lead to positive \( n_2 \). But since most materials expand when heated, the thermal nonlinearity leads to negative \( n_2 \). Under appropriate conditions other nonlinear interactions such as stimulated scatterings and multiphoton absorption may enter the picture. Although in any given experiment with high-power laser beams, all of these nonlinearities may be present, the experimental conditions can be adjusted such that only one or two of these effects have a dominant role in the measurements. It is a partial goal of this study to underline the microscopic processes involved in the observed nonlinearities. Unique characteristics of each process can be utilized to identify the dominating nonlinearity. Among these are polarization dependence of reorientational Kerr effect, temperature dependence of electrostriction, and intensity dependence of thermal effect and Brillouin scattering. Experiments were specially designed and performed in order to distinguish the strong from the weak. A detailed account of these experiments and their results with the description of the theory
and the relative significance of some of these nonlinearities will be carried out in Chapter IV.

As previously stated, no dispersion in $n_2$ was observed in the range of 0.5 to 1 $\mu m$ by previous workers.$^6,^7$ The results of the present work however indicates a relatively large dispersion in $n_2$ from 9.6 $\mu m$ to 10.6 $\mu m$. The variation of $n_2$ in this wavelength region is approximately a factor of four with the maximum of $(11.6 \pm 2) \times 10^{-10}$ (esu) at 10.24 $\mu m$. In addition, the $n_2$ measured in this work is an order of magnitude larger than that reported for the visible. The larger value of $n_2$ in this work is accounted for by the presence of electrostriction. But the presence of dispersion cannot be explained only in terms of electrostriction or Kerr effect. These issues will be discussed in Chapter V.

Finally, the measurements of $n_2$ were repeated using two other techniques. Power limiting was first tried. This method has been widely used, and is described in greater detail in reference 6. It is essentially another way to determine the critical power for self-focusing. The intensity on the axis in the far field transmitted through the nonlinear material is monitored as the incident power is increased. Above the critical power for self-focusing the on-axis transmitted fluence does not increase with incident power. Nonlinear refraction as well as the plasma generated by material breakdown limits the on-axis transmitted fluence.
This marks the onset of self-focusing. The second method was to measure the beam profile transmitted through the material. The beam shape is then fitted to the numerical solution of the nonlinear wave equation with $n_2$ as an adjustable parameter. It should be noted that in this method knowledge of the critical power for self-focusing is not important. The results of these two methods are in good agreement with the photoacoustic method. These results show that the photoacoustic method is a reliable and accurate method to measure $n_2$ and the formula for the critical power with its numerical constant given by Marburger is correct. This last point is important because the numerical constant in this formula used by some workers$^{15-17}$ was smaller by approximately a factor of four.
CHAPTER BIBLIOGRAPHY


CHAPTER II

EXPERIMENTAL APPARATUS

The experimental arrangement used for the measurements of $n_2$ in CS$_2$ is schematically shown in figure 2.1. It basically consists of a CO$_2$ TEA oscillator-amplifier system, an attenuator, beam steering optics and focusing lenses, a sample cell, and detectors. The laser beam is focused in the center of the CS$_2$ cell by lens L1. The transmitted light is collected into the detector number three by lens L2. This detector monitors the peak on axis fluence. The signal on D1 is used to trigger the digitizer. Detector D2 is the energy reference, and D4 monitors the back scattered light. D5 is a piezoelectric transducer and its output is proportional to the photoacoustic signal in the CS$_2$ cell. A room temperature HgCdTe detector is used for D6 to monitor the pulse shape. During the experiments an IBM/AT microcomputer was controlling the lasers and the attenuator and captured the digitized output of all the detectors in its memory for later analysis. In the following sections of this chapter each component of the experimental setup will be described in detail.
Figure 2.1—The Experimental Setup
The Master CO$_2$ TEA Laser

The heart of the system is a hybrid CO$_2$ TEA laser model PRF-150 from Laser Science, Inc. It consists of a high voltage discharge chamber at three torr above atmospheric pressure. This is only to ensure a constant flow of the gas mixture out of the discharge volume. The solid nickel electrodes are approximately 22 cm long and 3 cm wide separated by 2 cm and are highly polished. The laser is capable of firing at repetition rates of up to 150 Hz. To avoid the build-up of thermal effects in the nonlinear material and because of other limitations which will be discussed in the next section, it was operated at only 3 Hz. The discharge chamber together with cooling radiator and fans are totally enclosed in a quarter-inch-thick aluminum cylinder, which is an effective way of keeping electromagnetic interference away from the electronic detection system. The enclosure is sealed by two ZnSe windows at Brewster's angle. The gas mixture consists of 20 percent CO$_2$, 27 percent N$_2$, and 53 percent He. The optical resonator is 3 m long, with an Aluminum master grating mounted on one end as the tuning element. A spherical mirror with 10 m radius of curvature and 85 percent reflectivity and antireflection coated on the outside was used as the output coupler. It was mounted on three piezoelectric transducers for fine adjustments of the length of the cavity. Because of the constant problem of
optical damage to the intracavity optics, mirrors of ZnSe, Ge and GaAs were tried. No attempt was made to systematically study the damage properties of these mirrors. Individual differences, imperfections in the coating and lack of data hinders one from making any intelligent assessment as to which material was more damage resistant.

The multi-mode operation of the laser was suppressed by including a low pressure cw gain cell inside the optical resonator. A tube with active length of 50 cm and inner diameter of 1.2 cm was used. The premixed gas consisted of 11 percent CO$_2$, 22 percent N$_2$, 67 percent He, and was kept at the total pressure of 10 torr. The gain in the cw discharge tube could be adjusted by varying the voltage and gas pressure. The tube was capable of lasing on its own; a maximum of six watts of cw power at 10.6 μm (P20) was measured. For the single mode operation of the TEA laser, however, it was sufficient for the cw gain cell to operate below lasing threshold. In the hybrid TEA system the spectral dependence of the unsaturated gain is enhanced at line-center due to the presence of the low pressure gain cell. Single mode operation is achieved by placing an axial mode of the resonator at the line center. This single mode enjoys the benefit of the enhanced gain, and therefore dominates other modes in the cavity. This requires a fine adjustment of the resonator length within a fraction of a wavelength.
Piezoelectric ceramics were used to move the output coupler in a range of zero to six microns. The hybrid configuration was chosen over the injection locking method because a stable operation in the latter requires two resonators stable to within a fraction of the wavelength. In essence, the method of injection locking is quite straightforward; light from an external (usually cw) laser is used to "seed" the TEA CO₂ laser during the gain build-up. Very little injected power (<< 1 W) is needed for the single mode operation. A number of different methods have been successfully implemented for the actual injection of the external light into the cavity.¹,² Figure 2.2 shows a schematic representation of the hybrid and injection-locked configuration. In the injection-locking method active stabilization techniques must be used for cw and TEA resonators to keep both lasers on the same spectral line for long periods of time, which is tedious and expensive. On the other hand, the hybrid method offers the advantage that the output of the cw gain cell can be used as a reference for closed loop frequency stabilization. A complete description of the injection-locking method can be found in reference 3, and a good comparison with the hybrid technique is given in reference 4. Another advantage of the hybrid technique is the ability to vary the TEA laser pulse duration with the gain in the cw section. Figure 2.3 shows two pulse shapes with the cw gain cell operating (a) above
Figure 2.2—Single Mode Operation of CO\textsubscript{2} laser (a) Hybrid and (b) Injection Locking Configuration.
Figure 2.3—CO₂ Pulse Shapes (a) with 0.5 watts of CW power in the cavity and (b) with CW section operating below lasing threshold.
and (b) below lasing threshold. The pulse shapes are obtained using a room temperature HgCdTe detector with a response time of 1 ns. The signals from the detectors were displayed directly on a Tektronix oscilloscope model 7834 with a horizontal bandwidth of 500 MHz. When the cw laser is operated below threshold the TEA laser pulse has a width of 270 ns (FWHM). With 0.5 watts of cw power in the cavity the pulsewidth is 1.5 μs long (FWHM). The energy content of the pulse is 40 mj. It should be mentioned that no detectable change in the pulse energy was observed as a function of the gain in the cw discharge tube. For some experiments where a large peak power is not required operation of the gain cell above lasing threshold may be desirable. For the purpose of the study of the nonlinear refraction in CS$_2$ however the peak power in the 1 μs pulse was not sufficient. Also the heating problem posed by the presence of the cw power would severely affect the results of the experiments by way of creating a thermal gradient in the index of refraction and, in the long run, convective currents in the liquid CS$_2$. For these reasons, in the course of this study, the cw discharge tube in the laser was operated below lasing threshold.

In the study of the nonlinear refractive properties of the materials with high-power lasers, the knowledge of the intensity profile of the beam is crucial for a meaningful interpretation of the experimental results. It is essential
to suppress the multi-transverse mode operation of the laser because the beam profiles can be quite complicated. In the case of CS\textsubscript{2} where self-focusing is the dominant nonlinearity the use of multi-mode beams can lead to small scale self-focusing. To avoid the unnecessary burden of having to solve the nonlinear wave equation for a complicated beam pattern the laser was forced to operate in the TEM\textsubscript{00} mode. Ordinarily this can be accomplished by placing a diaphragm inside the resonator. This introduces more loss to the higher order modes in the resonator thus gives a winning edge to TEM\textsubscript{00} in the mode competition. In these experiments the small (1.2 cm) bore of the cw discharge tube played the role of the diaphragm inside the laser cavity. The single transverse mode operation of the laser was checked on a regular basis before each experiment. Figure 2.4 shows a beam profile obtained by a pinhole beam-scan. The solid line is a least squares fit of a Gaussian function to the experimental data points. The stability of the laser is evident from the small scatter of the data. The beam scan is performed just before the focusing lens L\textsubscript{1} and the half width at 1/e of the maximum is 3.7 mm. This gives $\omega_0 = 5.2$ mm ($\text{HWL}/e\text{M}$ of E). From this data the good quality of the optics used in the setup is evident. This is because the beam scan was performed after the beam had propagated through the entire experimental setup.
Figure 2.4—CO$_2$ Beam Profile. Data points are obtained with a 200 μm pinhole scan, and the solid curve is a least squares fit of a gaussian function.
The Slave Amplifier

The amplifier is a Lumonics kit model K-921. This is essentially a TEA discharge module without any resonator. Its electrodes are 47.5 cm long, 3 cm wide and 2 cm apart with rough surfaces. The discharge volume is enclosed inside a plexiglass box with two Brewster mounts on each end. A double-wall, fine copper mesh enclosure was constructed in order to protect the electronic equipment and detection system from electromagnetic interference. The two electrically isolated shields are connected to a common ground. This Faraday cage works remarkably well considering the huge noise generated by each burst of 40 KV along the 50 cm long electrodes. This is demonstrated in the small scatter of data. The power supply of the amplifier was not designed for delivering energy to the electrodes at a rate of more than 3 Hz. This was the main limitation on the repetition rate of the system.

Aside from the operational conditions of the amplifier, the most critical parameter for efficient energy extraction is the relative timing of the discharge between the master and the slave. An electronic trigger generator with adjustable delay was used to fire both lasers. Measurements of gain for 10.6 \( \mu \)m radiation were performed as a function of delay. Figure 2.5 shows the results for two different pulse shapes. Data points in (a) are for 1.5 \( \mu \)s long pulses of
Figure 2.5—Measurements of the gain in the amplifier at 10.6 μm (a) with CW section operating above and (b) below lasing threshold.
Figure 2.3 (a) and data points in (b) are for 270 ns pulses of figure 2.3 (b). The gain was calculated after reference 5 from

\[ E_{\text{out}} = E_{\text{s}} \ln \left(1 + \exp \left(g_0 L \left[\exp \left(E_{\text{in}}/E_{\text{s}} \right) - 1\right]\right) \right) \ (J/cm^2) \]

where \( E_{\text{in}} \) and \( E_{\text{out}} \) are the input and output energy density of the amplifier, \( E_{\text{s}} = 200 \text{ mJ/cm}^2 \) is the saturation energy density, \( g_0 \) is the gain coefficient, and \( L \) is the length of the active medium. For the shorter pulses which were used in all the experiments the maximum gain is 0.0384 cm\(^{-1}\) and it is available approximately 1 \( \mu \)s after the oscillator has fired. For completeness the data for 1.5 \( \mu \)s long pulses is also given. The reason for the smaller gain in this case is the presence of the cw beam in the amplifier before and during the pulse arrival, which quenches the gain. The gas mixture used in the amplifier for these measurements was 26 percent CO\(_2\), 13 percent N\(_2\), 61 percent He. Other gas mixtures were tested in the amplifier. The measured values of gain for 10.6 \( \mu \)m radiation at optimum delay and the discharge voltages are given in Table I. Measurements performed on the amplified pulses showed that the operation of the amplifier does not alter the beam profile or the pulse shape.
TABLE I
MEASURED VALUES OF GAIN IN THE AMPLIFIER
AT OPTIMUM DELAY

<table>
<thead>
<tr>
<th>% CO₂</th>
<th>% N₂</th>
<th>% He</th>
<th>g₀ (cm⁻¹)</th>
<th>voltage (KV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>26</td>
<td>13</td>
<td>61</td>
<td>0.0384</td>
<td>35</td>
</tr>
<tr>
<td>17</td>
<td>20</td>
<td>63</td>
<td>0.0315</td>
<td>36</td>
</tr>
<tr>
<td>20</td>
<td>27</td>
<td>53</td>
<td>0.0370</td>
<td>36</td>
</tr>
<tr>
<td>20</td>
<td>20</td>
<td>60</td>
<td>0.0383</td>
<td>36</td>
</tr>
</tbody>
</table>

The Attenuator

There are a number of different methods of continuous attenuation of a laser beam without deteriorating its quality. An ideal attenuator does not change the polarization state, the direction of propagation, and the intensity distribution of the beam. At the same time it offers large extinction ratio, high optical damage threshold, and a wide dynamic range in terms of the wavelength of the light. Ease of operation and the cost should also be considered in selection of an attenuator. A combination of a half wave plate and a polarizer has many of these properties. The half wave plates are used in normal incidence; they are not bulky and are easily rotatable. But a half wave plate can only be made for a single wavelength. A Babinet Soleil compensator can be used instead of the half wave plate. This is essentially a birefringent crystal (CdS) with adjustable
thickness. At any wavelength in the range of 9 to 11 μm this device can be tuned to be a half wave plate. Because these crystals are hard to grow in big pieces this expensive device has a very small (10 mm) aperture. The CO₂ beam must be condensed by means of a telescope. This exposes the compensator to optical damage.

Gaseous absorption cells have been used by other researchers. The variable attenuation can be achieved by bleeding an absorptive gas in a cell. For high power applications the nonlinear refractive effects in the gas cell may introduce more complications. A stack of rotatable Brewster plates can be used in transmission. In this case multiple reflections and the fact that the two faces of each plate are not perfectly parallel deteriorate the beam quality. Also in order to achieve reasonable extinction ratios four to six plates should be used which makes it an expensive device. Neutral density filters of CaF₂ were available, but were only used in front of detectors because of aberration and beam walkoff.

Initially a pair of wire grid polarizers were used in series. Wire grid polarizers have been commonly used for long wavelength applications, because the required wire spacing could be achieved without complications. For 10 μm applications, however, submicron wire spacings are required. Only recently the technology has become cost effective to
fabricate such fine structures. Aluminum wires were deposited on a grooved ZnSe substrate with 0.83 μm spacing. Both faces were antireflection coated. This structure reflects the light with polarization parallel to the wires, and transmits the component with electric field perpendicular to the wires. Wire grids are thin (3 mm) and are used in normal incidence, therefore easily rotatable. The wire grid polarizers can be used with any wavelength much larger than the wire spacing as long as the transmission of the substrate permits. With an extinction ratio of 1 to 500, wire grids appear to be ideal for an attenuator. However, they turned out to have unacceptable optical damage threshold. Failing to be useful as attenuators, wire grids became the subject of a careful study. Their optical damage threshold and absorption were measured as a function of wavelength and polarization of the light. The results of these measurements, together with a simple theoretical model which successfully describes the observed variation of absorption with wavelength and polarization, are given in Appendix A.

As a solution to the attenuation problem, an attenuator was designed to satisfy all essential requirements. This attenuator was constructed from a pair of large (4 by 16 cm) germanium plates with a clear aperture of 3.8 cm. Figure 2.6 shows the schematic setup of the attenuator. The analyzer was fixed and the polarizer was rotatable. Since at
Figure 2.6—The Attenuator Setup
Brewster's angle the reflected light is purely S-polarized, both plates were used in reflection. Two copper mirrors were used in conjunction with the Ge plate such that the input and output of the device were coaligned. The copper mirrors and the polarizer plate rotate as a whole. In the configuration shown in figure 2.6, the attenuator is functional only if its input beam is vertically polarized. Unfortunately, the CO$_2$ laser output was horizontally polarized. The polarization of the light was rotated from horizontal to vertical orientation by reflecting the beam from two copper mirrors before the beam enters the attenuator. The germanium plates were fabricated with a wedged angle of 2 degrees. This was to ensure that the reflections from the back face of the Ge plate diverge from the optical axis after a short (1 m) propagation distance. Germanium was chosen because of its large (4.002) index of refraction which gives a reflectivity of 78 percent for the S-polarized light. The variation of the index of refraction of Ge in the range of 9 to 11 $\mu$m is small; and the reflectivity of the P-polarized light remains close to zero for small changes of the angle of incidence around Brewster angle. Consequently, the attenuator could perform quite satisfactorily in this wavelength range. Although this device has a maximum transmission of only 60 percent, its minimum transmission can in principal be zero. The measurements of extinction ratio were limited only by the
signal-to-noise ratio of the detection system, which was 1000 to 1. Because the energy of the beam is spread over a large area on the surface of the Ge plate, this device has a high optical damage threshold. On the other hand, since the beam is deviated from the optic axis and brought back on itself by three reflections, the mechanical axis of rotation of the attenuator must coincide with the optic axis. Moreover, the rotation of a large object free from wobble requires special attention in design of the mechanical parts. The critical alignment of the attenuator was achieved by using a Melles Griot 0.5 watt HeNe laser. The HeNe beam was first aligned with the mechanical axis of rotation without the Ge plate and copper mirrors inside the rotating base of the attenuator. This was done by using two pinholes made specially to fit in the apertures of the attenuator to define the axis of rotation to within 200 μm. After the HeNe was sent through these pinholes the beam was allowed to propagate for at least 8 m; then the position of the beam was marked. Now the Ge plate was positioned at Brewster angle inside the attenuator and the two copper mirrors were mounted on the base. They were adjusted to bring the HeNe back on the same spot through the pinholes. The alignment was tested by rotating the attenuator. The same HeNe laser beam was then used to align the CO₂ laser.
The Optics

Turning mirrors in the setup were uncoated copper polished to a flatness of 1/20 of the wavelength at 10 μm. Uncoated Silicon mirrors were also used satisfactorily. Reflectivity of these mirrors was greater than 99 percent. Brewster windows of NaCl for the cw gain cell, and KCl for the amplifier were used. Because the salt windows are hygroscopic, they were kept at a higher temperature with respect to the room temperature, using heat cables and heat lamps. This procedure increased the life time of the windows significantly; nevertheless the salt windows had to be replaced on a regular basis due to deterioration of surface quality caused by environmental effects. Beam splitters used in the setup were ZnSe, with antireflection coating at 10.6 μm on one side. This eliminates the problem of multiple reflections. Meniscus lenses designed to compensate for the spherical aberration were used for focusing the laser beam in the CS₂ cell. All the lenses used for the experiment were ZnSe with antireflection coating on both sides. Germanium lenses were also available; they were only used in front of the detectors in order to collect the beam on the active area of the detector. The CS₂ cell was made from stainless steel. The cell was sealed with two NaCl windows and teflon o-rings. All the beam control optics used in the setup had a clear aperture size of greater than 3.5 cm. In order to change the
polarization state of the laser light from linear to circular, a phase-retarding reflector was used. This silicon mirror with a thin dielectric coating is used at 45°. In reflection from this mirror, there is a 90-degree phase deference between the S and P component of the electric field. The coating of the mirror was designed to cause the 90 degrees phase change only at 10.6 μm. At 9 μm for example, the phase change is decreased to 70 degrees. The reflectivity of the mirror is greater than 99 percent. Obtaining circular polarized light requires having equal components of S and P polarized in the incident beam. Although the Babinet soleil compensator is ideal for this purpose because of its tunability, it could not be used because of its small clear aperture. Frequently optical damage presented a problem with the intracavity optics. These optics were regularly checked and replaced if damaged.

Detection System

The positions of the six detectors used in the setup were shown earlier in figure 2.1. Detectors one through four are pyroelectric elements with an amplifier. The sensitive elements are LiTaO₃ with a flat frequency response from 2 to 20 μm. These elements are not suitable for resolving the temporal profile of the CO₂ pulses with their frequency response of 3 MHz. However, they are ideal for measurement of
time-integrated energy or fluence. The active elements of D1 and D3 are 3 mm, whereas those of D2 and D4 are 9 mm. The larger elements were used for these detectors to collect all of the energy in the beam. The energy reference detector D2 was calibrated using an ED200 pyroelectric detector manufactured by GenTec. This is a 1-inch active area, factory-calibrated pyroelectric detector. The piezoelectric transducer was made from a Lead Zirconate-Titanate (PZT) cylinder firmly mounted in a stainless steel casing. The details of the construction, advantages and characteristics of the PZT transducer are described in references 6 and 7. The first peak in the acoustic signal appears 15 μs after the arrival of the optical pulse. This is approximately the time it takes for the sound wave to travel from the focal volume to the transducer in CS$_2$. This signal is sent to the acoustic sampler with an adjustable time window. The acoustic signal within the time window are integrated and sent to the digitizer. A typical photoacoustic signal and the relative position of the time window are shown in figure 2.7. The measurement of the acoustic signal in this work was performed with the time window adjusted on the first peak of the acoustic signal. The signals from all of the detectors were digitized and sent to an IBM/AT microcomputer. Meanwhile, the laser amplifier and attenuator were controlled programatically by the microcomputer. The trigger control
Figure 2.7—Photoacoustic signal and its relation to the sampling time interval.
devices and the data acquisition network are represented by a block diagram in figure 2.8. At the outset of the experiment the computer sends an enable signal to the synchronizer. The synchronizer, once enabled, generates the trigger signals with appropriate delays at its internal repetition rate, which can be selected on its front panel. Since the mechanical shutter is the slowest device, it is triggered first. It takes 25 ms for the shutter to open fully. It can be kept open from 1 ms to 10 s. The shutter is used only when the cw gain cell is operating above lasing threshold. This is to block the unwanted cw beam as much as possible. Otherwise the shutter is left out of the system. 25 ms after the shutter the CO$_2$ laser is triggered. One microsecond later separate trigger signals are sent to the amplifier, acoustic sampler, and image digitizer. The delay and width of the time window in the acoustic sampler should be adjusted by displaying and comparing the acoustic signal with the trigger signals on the oscilloscope.

The infrared television camera is an array of pyroelectric elements. The active area of the vidicon camera is 18 x 24 mm, and there are 250 x 245 pyroelectric elements in a two-dimensional array. The elements are made from Triglycine Sulfate (TGS). This ferroelectric material requires that the electric dipole moment within all elements be oriented in the same direction to produce maximum pyro-
Figure 2.8—The Data Acquisition Network
electric effect. The reorientation of the dipole moments by applying an electric field is referred to as poling. It takes approximately 35 seconds for the camera control unit to perform the poling function automatically. Because dipole moments in the ferroelectric materials disorient at temperatures above the Curie limit, the vidicon camera should not be exposed to extremely hot objects. The Curie temperature of TGS is 49°C, and its boiling point is equal to that of water. With these characteristics it is obvious that measuring the beam profile of the CO$_2$ beam requires extreme care to avoid permanent damage to the sensitive elements of the vidicon camera. As little as a few tens of microjoules of energy can be detected. To avoid saturation and damage to the camera, the CO$_2$ laser beam was attenuated, using up to 2 cm of CaF$_2$. The output of the camera is a standard video signal and can be displayed directly on a television monitor. The video digitizer, developed at NTSU, begins to digitize the video information when the synchronization signal from the line selector coincides with the trigger signal from the central synchronizer. Only six subsequent lines of the video frame after the synchronization signal are digitized and stored in the internal memory of the video digitizer. The only limitation in digitizing the entire video frame is the size of the internal memory of the device, which can be easily increased. The data in the local memory of the video
digitizer is read by the computer when it is ready. The signals of D1 through D5 are digitized and sent to the computer by the other digitizer shown in figure 2.8. The small reflection from the Brewster window of the cw gain cell is directed toward D1, and its output is used to trigger the digitization process. To avoid triggering the digitizer with the noise and to reject misfires of the laser, a lower bound was imposed on the signal of D1. If the signal on D1 was smaller than this discrimination level the digitization process would not be initiated and the data point would be rejected. With proper adjustment of this voltage gate, over 90 percent of the pulses could be accepted with a small standard deviation in the data.
CHAPTER BIBLIOGRAPHY


CHAPTER III

MEASUREMENTS OF n_2

In the previous chapter the experimental apparatus was described in detail, including generation, control and detection of the laser light. The present chapter will concentrate on the experimental procedures leading to the measured values of nonlinear refractive index of liquid CS_2. First, some background material will be presented on general properties of liquid CS_2. The experimental technique and the method of extraction of n_2 will be discussed, and the results of the measurements in the spectral range of 9 to 11 μm will be presented. The magnitude of the errors in the measurement will be estimated and their main sources will be examined. Validity of the basic assumptions is experimentally verified. The motivation for these supporting experiments with their results will be considered in this chapter. Finally, the summary of the experimental results will make way for a theoretical analysis in the next chapter.

Carbon Disulfide

Carbon disulfide is a linear symmetrical molecule with zero dipole moment. At room temperature it is a colorless liquid dielectric. In the visible, pure CS_2 has a very small
absorption of 0.002 cm$^{-1}$; however, a much larger linear absorption is present in the infrared. Figure 3.1 shows the transmission of 2.2 cm of liquid CS$_2$ obtained by a Fourier transform spectrophotometer. Spectrophotometric grade CS$_2$ from Aldrich Chemical Company (940 W. Saint Paul Ave., Milwaukee, Wisconsin 53233) was used in all the measurements in this work. The minimum absorption of 0.11 cm$^{-1}$ occurs at 10.24 \( \mu \text{m} \). Two very large absorptions occur around 11 and 6 \( \mu \text{m} \), and a relatively large absorption of 0.6 cm$^{-1}$ is present at 9.6 \( \mu \text{m} \). The infrared absorption spectrum of CS$_2$ can be explained completely by the three fundamental vibrations of the molecule and their combinations. The two strongest infrared bands are at 396.7 and 1523 cm$^{-1}$ (25.2 and 6.5 \( \mu \text{m} \)); they are therefore to be interpreted as the two infrared active fundamental vibrations \( \nu_2 \) and \( \nu_3 \).\(^1\) \( \nu_2 \) is the vibrational bending mode and \( \nu_3 \) is the asymmetric vibrational stretching mode of the molecule. Since during the vibrations \( \nu_2 \) and \( \nu_3 \) the polarizability is the same at opposite phases of the motion, for small vibrations the polarizability does not change. Therefore these two modes are not Raman active, and do not appear in the Raman spectrum of CS$_2$.\(^2\) On the other hand the strongest Raman line of liquid CS$_2$ corresponds to 656.5 cm$^{-1}$ (15.2 \( \mu \text{m} \)),\(^1\) and in all probability this is the third fundamental \( \nu_1 \). \( \nu_1 \) is the symmetric vibrational stretching mode of the molecule. During the vibration of \( \nu_1 \)
Figure 3.1--Transmission of 2.2 cm room temperature liquid CS$_2$. 

Wavelength (μm)
the dipole moment remains zero; therefore $\nu_1$ is not an infrared active mode and does not appear in the infrared spectrum. The polarizability of the molecule, however, changes during $\nu_1$ vibration. This mode therefore is a Raman active mode and occurs in the Raman spectrum of CS$_2$. Figure 3.2 shows the vibrational levels of the ground electronic state of CS$_2$. The observed Raman and infrared transitions as reported in reference 2 are also indicated. All other absorption peaks of the infrared and the Raman spectrum are combination vibrations and overtones of the three fundamental modes of the molecule. And of course each vibrational level is split to many rotational levels. In general the rotational motion and the vibrational motion of the molecule are coupled together. In the region of interest in this study, which is the spectral range of CO$_2$ laser lines (9 to 11 $\mu$m), the two strongest absorptions occur at 11.6 and 9.6 $\mu$m. These two lines correspond to $\nu_3 - \nu_1$ and $\nu_1 + \nu_2$ combination vibrations respectively (855 and 1040 cm$^{-1}$). A dispersion in the linear index of refraction is therefore expected near each one of these absorption bands. Interferometric methods have been used$^3$ to measure the dispersion of CS$_2$ in the 2-15 $\mu$m region. The results of these measurements indicate a large dispersion of more than 30 per cent associated with the most energetic fundamental vibration $\nu_3$ at 6.5 $\mu$m. These measurements clearly show
Figure 3.2—Vibrational levels of the ground electronic state of liquid CS₂. The dashed lines are the infrared transitions and the solid lines are the Raman transitions.
dispersion of refractive index due to the other absorption bands. In particular dispersion, because of the strong nearby absorptions at 11.6 and 6.5 μm, are very close to the spectral region of interest in this work. However, the linear index of refraction throughout this wavelength range shows the small variation of only 0.7 per cent from its value of 1.626 at 10.6 μm.

The nonlinear optical properties of liquid CS$_2$ have been widely studied in the visible and near-infrared. In fact CS$_2$ is often referred to as a standard nonlinear material. Because of the inversion symmetry this material does not exhibit any second order nonlinearity. But a variety of nonlinear effects based on third order susceptibility have been observed under a wide range of experimental conditions. Among these nonlinear effects are self-focusing, stimulated Brillouin scattering, stimulated Raman scattering, optical phase conjugation, optical Kerr effect, ellipse rotation and intensity induced birefringence and nonlinear absorption. All of these effects have been studied in the visible. The extensive study of the nonlinear properties of liquid CS$_2$ in the visible is not nearly matched in the infrared region of the spectrum. However, the possibility of using CS$_2$ as an infrared ac Kerr switch was demonstrated early after the invention of atmospheric pressure CO$_2$ lasers. In these
experiments the reorientational Kerr effect in the CS$_2$ was used. A CO$_2$ laser pulse induced optical birefringence for a visible probe. The state of polarization of the probe therefore depended on the intensity of the infrared beam. The visible beam could be switched on and off through a pair of crossed polarizers by the CO$_2$ laser pulse. Owen$^{24}$ estimated the value of the nonlinear refractive index of CS$_2$ at 10.6 $\mu$m by performing just such an experiment. To my knowledge the only other measurement of $n_2$ in CS$_2$ at 10.6 $\mu$m is done by Jungling.$^{25}$ He estimated the value of the critical power for self-focusing by observing the distortions of the beam shape after propagating through the CS$_2$. Jungling then calculated a value for the nonlinear refractive index which was three orders of magnitude larger than that estimated by Owen. In light of the developments that will be described in this and the next chapter, some aspects of this disagreement will be understood.

The Photoacoustic Method

When an intense laser pulse propagates through a liquid, a pressure wave is generated which propagates outward from the beam volume at the speed of sound. Diverse processes can be responsible for the generation of this opto-acoustic signal. Some of the possible opto-acoustic generation mechanisms, in the order of increasing efficiency are electrostriction, thermal expansion, photochemistry,
molecular dissociation, bubble formation, and material breakdown. The theory of the opto-acoustic generation has been worked out in some length, and the technique has found many applications from measurement of weak absorptions to material testing. In this study the photoacoustic signal was used to measure the critical power for self-focusing. A piezoelectric transducer was coupled to the walls of the liquid CS₂ cell to detect the opto-acoustic signal in the experiment. The details of the detection system and the experimental setup were discussed earlier in Chapter II. The CO₂ laser beam was focused sharply in the center of the 2.2 cm long CS₂ cell by lens L1 as shown in figure 3.3. The optoacoustic signal was then monitored as a function of increasing incident power of the pulse. A high sensitivity of opto-acoustic detection at the threshold of the dielectric breakdown was expected owing to the large increase in the conversion efficiency for this case over all the other acoustic signal sources. Figure 3.4 shows the results of the photo-acoustic signal up to the threshold for dielectric breakdown at 10.6 μm. The dramatic increase of the acoustic signal is observed as an abrupt steepening of the signal at the onset of breakdown. The focal length of the lens L1 was 6.35 cm, this gives a spot size at the focus of 27 μm (HWHM of intensity). To show that in CS₂ the optical breakdown is self-focusing-initiated, the measurement
Figure 3.3—The photoacoustic setup and the piezoelectric transducer.
Figure 3.4—The photoacoustic signal versus incident power at 10.6 µm.
was repeated with a different focal length lens Ll. A lens with focal length of 12.7 cm was used. This varied the irradiance by a factor of four for a given input power. The results, presented in figure 3.5, clearly show that the onset of optical breakdown depends on the input power rather than on input irradiance. At the critical power for self-focusing the irradiance increases rapidly as the light propagates in the nonlinear material. Therefore the material breakdown can occur even if the calculated incident intensity inside the medium without self-focusing is smaller than the breakdown threshold. As was mentioned in Chapter I, the change of the index of refraction responsible for self-focusing is irradiance-dependent. Whole-beam self-focusing results when the irradiance-dependent self-lensing overcomes diffraction. However, for a given input power, both the irradiance and the effect of diffraction decrease as the focal area increases. The net result is that the power required for the self-lensing to overcome diffraction (i.e., the critical power for self-focusing) remains constant as the focal area changes.\textsuperscript{34-37}

Other nonlinear processes, such as stimulated Brillouin scattering, could in principle, be the mechanism responsible for the large threshold like increase in the acoustic signal shown in figure 3.5. However, unlike self-focusing, the onset of the stimulated Brillouin scattering is irradiance-
Figure 3.5a--The photoacoustic signal versus incident power at 10.6 μm for two lenses. The circles are for L1=6.35 cm, and the squares are for L1=12.7 cm focal-length lenses.
Figure 3.5b--The photoacoustic data of Fig. 3.5a plotted versus incident intensity for two lenses. The circles are for $L_1=6.35$ cm, and the squares are for $L_1=12.7$ cm focal-length lenses.
dependent. The irradiance threshold for the onset of stimulated Brillouin scattering will be calculated based on a steady-state theory in the next chapter. The intensity at the focus of the long-focal-length lens was not large enough for stimulated Brillouin scattering to occur. In order to observe stimulated Brillouin scattering, high intensities and long interaction lengths are needed. The scattered light from the end of the interaction region propagates backward and gains energy from the forward-traveling pump beam. The effective gain length for this nonlinear process in the case of sharply focused beam is approximately equal to the Rayleigh range, which is the region of high intensity. At incident powers above the critical power for self focusing the gain length for the stimulated Brillouin scattering is effectively equal to the self-focusing length, which is even shorter than the Rayleigh range. This does not permit the back scattered light to build up. In all the measurements performed, the detector D4 (figure 2.1) monitored the back-reflected light from the nonlinear material. Neither increased backscattered nor pump depletion characteristic of stimulated Brillouin scattering was observed. These and the fact that the onset of the sharp increase in the acoustic signal is independent of the focal volume, confirm that the large acoustic signal shown in figure 3.5 is initiated by self-focusing.
Extraction of $n_2$.

In the previous section it was established that the large intensity required for the material breakdown was provided by the catastrophic collapse of the beam due to self-focusing. Therefore we can directly read the threshold power from figure 3.5 to be $160 \pm 32$ kW. The greatest source of uncertainty in the measurement of the peak power was the shot-to-shot variation of the energy content of the tail of the CO$_2$ laser pulse. This, in large part, is responsible for the $\pm20$ per cent absolute error in the measurement of the peak power in these experiments. The threshold power can be determined from photoacoustic data based on the abrupt increase in the scatter of data at the onset of self-focusing. Since each data point is the average of many laser shots, monitoring the standard deviation of each data point provides a uniform method of determination of this threshold power.

In the absence of any linear absorption this incident threshold power is the same as the critical power for self-focusing. At 10.6 $\mu$m the linear absorption coefficient is 0.21 cm$^{-1}$. This absorption reaches a minimum of 0.11 cm$^{-1}$ at 10.24 $\mu$m. Under these conditions the critical power for self-focusing is not exactly equal to the threshold power required for the onset of the large acoustic signal. As was pointed out earlier, the nonlinear refractive index
responsible for self-focusing depends on the intensity of the beam. Under sharp-focusing geometry the high intensity required for a significant refractive index change is achieved only at the focal area. Therefore it is reasonable to assume that as the incident power is increased the greatest nonlinear refraction first occurs at the position of highest intensity (i.e., the Rayleigh range). It should be stressed that the Rayleigh range is also the region in which the strongest diffraction occurs. Then if the power delivered to the focal volume is equal to the critical power for self-focusing, the nonlinear change of the refractive index overcomes the linear diffraction effect and the beam collapses. The critical power for self focusing is therefore related to the threshold power according to Beer's law

$$ P_c = P_t \exp(-a\ell) $$

where $P_t$ is the power incident on the entrance face of the sample, $P_c$ is the power delivered to the focal volume, $a$ is the linear absorption and $\ell$ is the distance of the focus from the front surface of the nonlinear cell. The effect of the linear absorption on the nonlinear propagation will be discussed in more detail in the next chapter and the correctness of the assumption in this argument will be verified experimentally. At this point, however, we calculate the critical power for self-focusing at 10.6 μm to be $127 \pm 25$ kW, based on the relation given above. Using
Marburger's\textsuperscript{34} formula, which was discussed in Chapter I, \( n_2 \) can be directly calculated. Therefore the \( n_2 \) for CS\textsubscript{2} at 10.6 \( \mu \)m is evaluated to be \((3.13 \pm 0.7) \times 10^{-10}\) esu. This value compared to the value of \( 1.3 \times 10^{-11} \) esu measured in the visible\textsuperscript{7} and near-infrared is larger by an order of magnitude.

**Measurement at Other Wavelengths**

Up to now we have developed an experimental method and a procedure to evaluate the nonlinear refractive index. At this point we will apply this technique to measure \( n_2 \) of CS\textsubscript{2} at some other strong lines of the CO\textsubscript{2} laser. The value of \( n_2 \) at 10.6 \( \mu \)m presented in the previous section suggests an order of magnitude dispersion. Further studies of the dispersion of \( n_2 \) near 10.6 \( \mu \)m should better our understanding of the microscopic mechanisms responsible for \( n_2 \). In reference 7 it was established, in broad agreement with the existing literature, that the value of \( 1.3 \times 10^{-11} \) esu for \( n_2 \) in the visible was caused by the reorientation of the molecules. It will be shown in the next chapter that the large increase in the value of \( n_2 \) from 1 to 10 \( \mu \)m should not be interpreted completely as due to dispersion, but partly because of the presence of other nonlinearities along with the reorientation of the molecules under the experimental conditions.

The CO\textsubscript{2} laser was tuned by changing the angle of the grating with respect to the optic axis. As expected, this did
not alter the mode quality of the beam, or the pulse shape. The results of measurement at 10.55 μm are shown in figure 3.6. These results were obtained using two different focal length lenses of 6.35 and 12.7 cm. These results clearly show that the nature of the nonlinearity at 10.55 μm is the same as that observed at 10.6 μm. The threshold power for self-focusing, however, is smaller by approximately a factor of two. The linear absorption at 10.55 μm is 0.19 cm⁻¹, which is only 10 per cent smaller than absorption at 10.6 μm. Therefore the decrease in threshold power at 10.55 μm cannot be completely explained by the effect of linear absorption. As a result the critical power for self-focusing varies by a factor of approximately two in the small wavelength range of 10.6 to 10.55 μm; this suggests a large dispersion of nonlinear refractive index in this spectral range.

We continue to decrease the wavelength, and figure 3.7 shows the results of measurement of the photoacoustic signal versus incident power at two wavelengths of 10.5 and 10.3 μm. Clearly the strength of the nonlinearity continues to grow as the wavelength is decreased. The threshold power at 10.27 and 10.24 μm decreases further. These results are shown in figure 3.8. The next intense line of CO₂ laser where a measurement was performed is 9.6 μm. At this wavelength, however, the linear absorption of CS₂ has increased to 0.6 cm⁻¹. The results of this measurement are shown in
Figure 3.6—The photoacoustic signal versus incident power at 10.55 μm for two lenses. Points are for L1=6.35 cm and circles are for L1=12.7 cm focal-length lenses.
Figure 3.7--The photoacoustic signal versus incident power at two wavelengths. The points are for 10.5 μm, and the circles are for 10.3 μm.
Figure 3.8—The photoacoustic signal versus incident power at two wavelengths. The points are for 10.27 μm and the circles are for 10.24 μm.
figure 3.9. The output power of the laser was not large enough to reach the onset of the nonlinearity. The measurement at this wavelength, therefore, provides only a lower bound on the magnitude of the nonlinear refractive index. The results of the measurements of the threshold power together with the calculated values of the critical power for self-focusing and the nonlinear refractive index are shown in Table II and figure 3.10. In the remaining sections of this chapter two other methods of measurement of $n_2$ will be discussed, and their results will be compared with the values of $n_2$ obtained by the photo-acoustic technique.

### TABLE II

THE PHOTOACOUSTIC MEASUREMENTS OF $n_2$

<table>
<thead>
<tr>
<th>$\lambda$ ($\mu$m)</th>
<th>$\alpha$ (cm$^{-1}$)</th>
<th>$P_t$ (kW)</th>
<th>$P_c$ (kW)</th>
<th>$n_2 \times 10^{10}$ (esu)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10.6</td>
<td>0.21</td>
<td>160 ± 32</td>
<td>127 ± 25</td>
<td>3.13 ± 0.6</td>
</tr>
<tr>
<td>10.55</td>
<td>0.19</td>
<td>80 ± 16</td>
<td>65 ± 13</td>
<td>6.05 ± 1.1</td>
</tr>
<tr>
<td>10.5</td>
<td>0.17</td>
<td>60 ± 12</td>
<td>50 ± 10</td>
<td>7.8 ± 1.5</td>
</tr>
<tr>
<td>10.3</td>
<td>0.12</td>
<td>38 ± 8</td>
<td>33 ± 7</td>
<td>11.3 ± 2</td>
</tr>
<tr>
<td>10.27</td>
<td>0.11</td>
<td>40 ± 8</td>
<td>35 ± 7</td>
<td>10.65 ± 1.9</td>
</tr>
<tr>
<td>10.24</td>
<td>0.11</td>
<td>36 ± 7</td>
<td>32 ± 6</td>
<td>11.6 ± 2</td>
</tr>
<tr>
<td>9.6</td>
<td>0.6</td>
<td>&gt;120</td>
<td>&gt;62</td>
<td>&lt;5.25</td>
</tr>
</tbody>
</table>

The Power-Limiting Technique

The power-limiting method was briefly discussed in Chapter I, and the experimental setup used for this measurement
Figure 3.9--The photoacoustic signal versus power at 9.6 μm. The output of the laser is less than the threshold of the nonlinearity.
Figure 3.10—Results of the measurements of $n_2$ using the photoacoustic method.
technique was described in Chapter II. The light transmitted through the CS$_2$ cell is collected into a pinhole using lens L2, as shown in figure 3.11. The diameter of the beam at the pinhole is approximately the same as that of the pinhole. The signal on detector D3 behind the pinhole is monitored as a function of the incident power. Figure 3.12 shows the results of power-limiting measurement at 10.6 μm. At high incident powers the light is no longer reimaged onto the detector D3 because of self-focusing. In addition, the plasma created by optical breakdown of the material blocks additional light from reaching the detector. These two effects limit the energy reaching the detector D3. This limiting occurs at the same input power that produces an abrupt increase in the optoacoustic signal. Typically, in optical power-limiting experiments, what is first observed is an increase in the scatter of data for the energy reading D3 near the critical value for the input power, as seen in figure 3.12. The threshold power for self focusing obtained by this method at 10.6 μm is 180 ± 30 kW. This and the result of the optoacoustic measurement have overlapping error bars, and therefore are in agreement. Figure 3.13 shows the results at 10.27 μm. The threshold power for limiting is 38 ± 8 kW, which is to be compared with the value of 40 ± 8 kW obtained by the optoacoustic method. The results presented in this section confirm that the photoacoustic
Figure 3.11—The power limiting setup. At low powers the beam is collected into the pinhole P.H. (solid line). With incident powers above the critical power for self-focusing the beam no longer is reimaged into the pinhole (dashed line).
Figure 3.12—Signal of $D_3$ versus incident power at 10.6 μm.
Figure 3.13—Signal of $D_3$ versus incident power at 10.27 μm.
technique is an accurate and reliable method. A third method of measurement of $n_2$, which without any doubt confirms the accuracy of the results as well as the role of self-focusing, is described in the next section.

Beam Distortion Measurement Method

Intensity-induced change of index of refraction in a Kerr like media, causes a nonlinear phase change in the beam during the propagation in the material. As already noted, if $n_2$ is positive and the self-focusing distance is smaller than the length of the sample, catastrophic intensity rise occurs. When the self-focusing length is longer than the length of the sample, the beam does not collapse. The nonlinear phase change, however, is imprinted on the beam. This affects the beam shape in the far field. The way in which the intensity profile and the phase profile interact through the material nonlinearity is of course governed by the nonlinear wave equation. The wave equation with its initial conditions was solved numerically. The numerical solutions were fitted to the measured beam profile in the far field using $n_2$ as an adjustable parameter. The fitted value of $n_2$ then is compared with that obtained by the optoacoustic method. It is not the intention of this section to catalog the rich behavior of the solutions of the nonlinear wave equation. A comprehensive account of the numerical computations and many different approximate analytical solutions to the
self-focusing equations are given by Marburger.\textsuperscript{34} The wave equation which is nothing but the nonlinear Schrödinger equation has been solved exactly,\textsuperscript{38} in its two-dimensional form, by reducing it to the inverse scattering problem for a certain linear differential operator. In this work, a predictor corrector method due to Hamming\textsuperscript{39} was used to solve the three-dimensional equation, assuming cylindrical symmetry and instantaneous response of the nonlinearity. The validity of the first assumption is clear because of the good quality of the beam. The discussion of the microscopic mechanism of the nonlinearity in the next chapter justifies the second assumption. Full description of the wave equation and the numerical method is given in Appendix B. Starting with the initial conditions, the wave equation is solved step by step to the end of the sample. From there to the position of the detector the beam propagates linearly. The beam profile at the detector is calculated by evaluation of the Fresnel integral. Since the detectors measure the time-integrated energy, the beam profile is time-integrated over the pulse shape. Because of the limitations in computation time, the pulse shape was approximated by two-step functions as shown in figure 3.14. The data points are obtained from six individual pulses. Detailed study of the pulse shape shows that, although the total variation of the energy of the pulse is only 6 per cent, the variation of the energy content of
Figure 3.14—The approximate pulse shape used in the numerical solutions of the wave equation (solid line). The points are measurements made by the fast HgCdTe detector, and the dashed line is a typical pulse shape.
the tail of the pulse is 15 per cent, whereas the variation of the energy of the peak of the pulse is 2 per cent. This is apparent from the greater scatter of the data points in the tail of the pulse. As was pointed out earlier, this is the greatest source of error in the calculation of the peak power. The approximated pulse shape is also shown in figure 3.14. The energy content of the tail of the pulse is one third of the total energy of the pulse. The maximum power in the tail, however, is only ten per cent of the peak power. So when the peak of the pulse is propagating non-linearly, the tail does not have enough power to induce any change in the index of refraction. One-third of the energy of the pulse therefore propagates linearly. This is true if the response of the material is instantaneous.

The beam profile was measured using the infrared imaging system. The experimental setup for these measurements is shown schematically in figure 3.15. The vidicon camera and its digitizer are described in Chapter II. A 25.4-cm-long focal length lens was used to focus the light in the center of the CS$_2$ cell. With this geometry the self-focusing distance was longer than the length of the cell, and therefore total collapse of the beam did not occur in the sample. This is desirable in these measurements, because of two reasons. First, the numerical computations do not converge at distances very close to the singularity, and
Figure 3.15—Setup for the beam distortion measurements. The position of the focus is at a distance 1 from the front surface of CS$_2$. 
typically they are not reliable beyond singularity. The second reason is a practical one, namely, the material breakdown following the collapse of the beam. The plasma generated in the focal volume at the peak of the pulse absorbs and scatters the trailing edge of the pulse. Under these conditions the simple third-order nonlinearity no longer is an adequate model of the interaction of light with the material. Therefore to avoid complications caused by the generation and propagation of the plasma, and its interactions with the light and the CS$_2$ molecules, as well as the acoustic shock waves, it is essential to avoid catastrophic intensity rise in the material.

The first measurement is performed at 10.27 $\mu$m. The beam profile at a distance of 20 cm behind the CS$_2$ cell is shown in figure 3.16. The profile is obtained by averaging ten laser pulses. The incident power is 15 kW, which is approximately three times smaller than the threshold power for self-focusing. This is the region of near-linear propagation. The beam profile computed from the wave equation is also shown in the same figure. When the incident power exceeds the threshold power for self-focusing some energy diffracts into the wings of the beam, as shown in figure 3.17. This data is obtained with an incident power of 40 kW. The solution of the wave equation is also plotted in this figure. The value of $n_2$ used in the computation of
numerical results of figure 3.16 and figure 3.17 is 9.8 x 10^{-10} esu, which is to be compared with the value of (10.65 ± 1.9) x 10^{-10} esu given in Table II. This remarkable agreement confirms once again the accuracy of the results and the method of indirect extraction of $n_2$ in the photoacoustic technique. It should be stressed that in the present method the role of the linear absorption is directly taken into account in the solution of the wave equation.

The fitting procedure is based on visual inspection and comparison, simply because of the limitations on the computational power available. Although this may be a relatively subjective mode of action, it is possible to determine realistic bounds on the accuracy of the eyeball fitting procedure. Numerical experiments show that the beam shape is a sensitive function of $n_2$. Although this sensitivity depends on the geometry and the initial conditions, as a general rule it was found that a 20 per cent variation of $n_2$ from its optimum value changes a good fit to an unacceptable fit. This is illustrated in figure 3.18. The experimental points are the same as that of figure 3.17, but for the calculated curve the value of $n_2=12 \times 10^{-10}$ esu was used.

The next set of beam shape measurements is performed at 10.5 μm. Figure 3.19 shows the beam shape in the far field with incident power of 55 kW. The focus was positioned inside the sample at a distance of $=1.4 \text{ cm}$ from the front
Figure 3.16—Beam shape measurement at 10.27 μm (dotted line). Incident power is 15 kW. The calculations (solid line) are performed with $n_2 = 9.8 \times 10^{-10}$ esu.
Figure 3.17—Measurement of the beam profile at 10.27 μm (dotted line). Incident power is 40 kW. The calculations (solid line) are performed with $n_2 = 9.8 \times 10^{-10}$ esu.
Figure 3.18—The beam shape is sensitive to the value of $n_2$. The data of Fig. 3.17 (dotted line) is fitted to the solution of the wave equation using 20% larger $n_2$ (solid line $n_2 = 12 \times 10^{-10}$ esu). The fit is unacceptable.
Figure 3.19—Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is 1 = 1.4 cm inside the CS₂. The calculated curve (solid line) is drawn by using $n_2 = 8 \times 10^{-10}$ esu.
A value of $n_2=8 \times 10^{-10}$ esu was used for the calculated beam profile shown in figure 3.19. The nonlinear propagation is sensitive to the position of the focus. This is mainly because of the interplay between linear absorption and the nonlinear refraction effects, and partly because of the initial phase of the beam. In order to study the effect of the position of the focus on the nonlinear propagation, the CS$_2$ cell was mounted on a translation stage. The position of the focus ($\ell$) was varied without any other changes in the experimental conditions of figure 3.19. This variation is from 1.4 cm inside the sample to 0.64 cm out in front of the sample. The whole range of movement of the cell is 2 cm, which is approximately equal to the Rayleigh length. By changing the position of the cell the input phase and intensity of the beam was changed, keeping the power constant.

Figures 3.20 through 3.25 show the results of these measurements. In each of these figures the calculated beam profile is also plotted. The values of $n_2$ used to calculate the fitted beam profiles are shown in Table III. The average value of $n_2$ in Table III is $(8.5 \pm 0.7) \times 10^{-10}$ esu which is to be compared with $(7.8 \pm 1.5) \times 10^{-10}$ esu given in Table II. Negative numbers for $\ell$ mean that the focus is out in front of the CS$_2$ cell. As seen in figure 3.25, with $\ell=-0.64$ the change in the beam shape is not as dramatic as for the cases where the focus is positioned inside the
Figure 3.20—Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is $l = 1.4$ cm inside the CS$_2$. The calculations (solid line) are performed using $n_2 = 7.2 \times 10^{-10}$ esu.
Figure 3.21—Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is $l = 0.38$ cm inside the CS$_2$. The calculation (solid line) is performed using $n_2 = 8.3 \times 10^{-10}$ esu.
Figure 3.22—Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is $l = 0.13$ cm inside the CS$_2$. The calculation (solid line) is performed using $n_2 = 8.3 \times 10^{-10}$ esu.
Figure 3.23--Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is $l = 0.0$ cm. The calculation (solid line) is performed using $n_2 = 9.2 \times 10^{-10}$ esu.
Figure 3.24—Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is 1 = -0.13 cm out in front of the CS₂ cell. The calculation (solid line) is performed using $n_2 = 9.5 \times 10^{-10}$ esu.
Figure 3.25—Measurement of the beam profile at 10.5 μm (dotted line). The position of the focus is \( l = -0.64 \) cm out in front of the CS\(_2\) cell. The calculation (solid line) is performed using \( n_2 = 9 \times 10^{-10} \) esu.
TABLE III
VALUES OF n\textsubscript{2} WITH DIFFERENT POSITIONS OF THE FOCUS OBTAINED FROM NUMERICAL COMPUTATIONS AT 10.5 \textmu m

<table>
<thead>
<tr>
<th>\ell (cm)</th>
<th>n\textsubscript{2} \times 10\textsuperscript{10} (esu)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.4</td>
<td>8</td>
</tr>
<tr>
<td>1.14</td>
<td>7.2</td>
</tr>
<tr>
<td>0.38</td>
<td>8.3</td>
</tr>
<tr>
<td>0.13</td>
<td>8.3</td>
</tr>
<tr>
<td>0.0</td>
<td>9.2</td>
</tr>
<tr>
<td>-0.13</td>
<td>9.5</td>
</tr>
<tr>
<td>-0.64</td>
<td>9</td>
</tr>
</tbody>
</table>

sample. This confirms the earlier assertion that the greatest nonlinear refraction occurs at the focus where the intensity is maximum. In addition to reinforcing the confidence in the photoacoustic method, this experiment shows that the model considered, namely the self-focusing equation including linear absorption, is adequate to describe the observed nonlinearity.

Conclusion

At this point we have reached the goal of this chapter, which is to present the results of measurements of n\textsubscript{2} in CS\textsubscript{2} for the spectral range of the CO\textsubscript{2} laser. All the measurements were performed at room temperature. General information on the properties of CS\textsubscript{2} was presented, which provides some insight on the nature of the interaction of
individual molecules with the infrared electromagnetic radiation. In this chapter the photoacoustic method was developed, and it was shown that by using this method accurate results can be obtained with relative ease. No sensitive alignment is required, as is the case for the power-limiting technique, and no extensive computation is called for, as is the case for the beam distortion measurement method. In addition, the method offers high sensitivity. Intuitive arguments and some evidence of support were presented in favor of the way in which the role of linear absorption was taken into account in extraction of $n_2$. This issue will be clarified in a direct experimental fashion in the next chapter.

Table II summarizes the results obtained by photoacoustic method. These results show that the $n_2$ in this region of wavelength is an order of magnitude larger than for reorientational $n_2$ measured in the visible and near-infrared. Up to this point no reference has been made to the microscopic mechanism responsible for the nonlinearity. It will be shown in the next chapter that the larger value of $n_2$ in these experiments is not all due to dispersion, but it is mainly due to the presence of electrostriction in addition to the reorientational nonlinearity. This is the case for the long pulses and small spot sizes used in these measurements. With the experimental methods employed in this
work it is not possible to distinguish directly between the reorientational nonlinearity and the electrostriction effect. The results obtained using these methods reflect the sum total of both nonlinearities. In the next chapter, however, the relative strength of these two effects will be estimated by considering the polarization dependence of the nonlinearities.

The data presented in Table II indicate the presence of a large dispersion. Thermal defocusing could in principle cause an apparent dispersion in the nonlinear refractive index. However, the role of thermal self-defocusing is negligible. Experimental evidence supporting this point is presented in this chapter in figures 3.5a and 3.6. It is already pointed out that the critical power remains constant when the intensity changed by a factor of four. Thermal self-defocusing on the other hand is intensity-dependent. These two experiments rule out the presence of thermal effects. The possible sources of the observed dispersion will be considered in Chapter V, where the dispersion characteristics of reorientational and electrostrictive nonlinearities in the neighborhood of vibrational resonances will be discussed. In the remainder of this chapter the two well known methods of measurements of $n_2$ were described. The results obtained by the photoacoustic method were verified by power-limiting and beam shape measurement techniques. The
agreement among these three methods is remarkable. In particular the beam distortion measurement not only confirms the presence of dispersion but verifies that a steady-state self-focusing model is adequate to describe the nonlinearity.
CHAPTER BIBLIOGRAPHY


CHAPTER IV

PHYSICAL MECHANISMS

The results of the measurements of the nonlinear refractive index of \( \text{CS}_2 \) using three different methods were presented in Chapter III. In arriving at these results the steady-state self-focusing model was used to interpret the experiments. It was experimentally shown that this was an appropriate choice. However, the physical mechanism responsible for the observed self-focusing was not considered. In the present chapter the discussion of the microscopic mechanisms relevant to the experimental conditions will be presented. It has already been pointed out that the reorientational Kerr effect is shown\(^1,2\) to be the dominant nonlinear mechanism in \( \text{CS}_2 \) at 0.5 and 1 \( \mu \text{m} \). The results in the previous chapter indicate that \( n_2 \) increases by an order of magnitude when the wavelength is changed from 1 to 10 \( \mu \text{m} \). It will be shown that this increase is because of the presence of electrostriction under our experimental conditions. In section one of this chapter the strength of the Kerr effect will be compared to the electrostriction. This question will be further studied experimentally. The temperature and polarization dependence of these two effects will be used to distinguish the dominant mechanism. As was discussed in
Chapter III, no experimental evidence for the presence of stimulated Brillouin scattering was observed. In section two of this chapter this phenomenon will be considered and the intensity threshold for the stimulated Brillouin scattering will be calculated. As seen in Chapter III, a large dispersion in \( n_2 \) exists. The microscopic mechanism of this dispersion is not rigorously understood. It is apparent that \( n_2 \) increases with the decrease of linear absorption. This could possibly be explained by the thermal contribution to the change of refractive index. The negative contribution of the thermal effect to the nonlinear index of refraction is linked to the linear absorption. Therefore, if the thermal effect is significant, increasing the linear absorption would increase the thermal defocusing, which in turn increases the self-focusing threshold. This would lead to an apparent dispersion, since the value of the power threshold for self focusing is used to calculate \( n_2 \). But under experimental conditions the thermal effect was not significant. Experimental evidence to support this point is given in Chapter III. It is shown that the power threshold for the onset of nonlinearity measured at more than one wavelength is independent of the focal volume. Thermal contribution to the change of the index of refraction should increase as the focal volume decreases. Measuring the same power threshold for self-focusing, using lenses of two different focal
lengths, however, shows that the thermal contribution to $n_2$ is smaller than the error bars in the measurements. In section three of this chapter the contribution of thermal variation of the index of refraction will be examined. It will be shown that variation of the refractive index with temperature is too slow to be significant during the length of the CO$_2$ pulse. Furthermore, the effect of linear absorption on self-focusing will be studied in a separate set of experiments. The discussion of these experiments and their results will be given in section four of this chapter. These results confirm the accuracy of the indirect method by which the role of linear absorption was taken into account in the photoacoustic method.

Comparison of The Reorientational Kerr Effect and The Electrostriction

When an intense linearly polarized light pulse travels through an optically isotropic medium, the material becomes anisotropic. One mechanism responsible for this effect is the reorientation of the molecules. In particular it produces a large effect in fluids with anisotropic molecules. For the infrared radiation in an anisotropically polarizable molecule the electronic contribution to the optical Kerr effect is negligible compared to the reorientational contribution. The intense electric field induces a dipole moment in the molecule. For a long molecule
with cylindrical symmetry, such as CS₂, the induced dipole moment is in the direction of the axis of symmetry of the molecule. This is because the polarizability is much greater in the long direction of such molecules. The incident field exerts a torque on the molecule through the interaction with the induced dipole moment. Because of the inertia, the molecules cannot follow the field at the high optical frequency. The torque, however, has a component which varies slowly in time (i.e., the field envelope). This tends to align the molecules in the direction of the field, which results in a birefringence due to the inherent anisotropy in the polarizability of the molecules. For CS₂ it has been demonstrated that the molecules respond with an ultrafast component superimposed upon a slower relaxation of molecular orientation. The fast librational decay time is measured to be 0.24 ps. During this time, superimposed on their random thermal motion they have a "coherent" rotational motion in phase with the driving field. Once the field is turned off, this phase memory will decay. Individual molecules need to move a small fraction of the total path toward eventual rotational randomization to have a large macroscopic effect. This relaxation time, during which the birefringence disappears and thermal random motion takes over, has been measured to be 2.1 ps for liquid CS₂. Such fast response times compared to the pulse durations used in
these experiments can be considered instantaneous. The freedom of rotation in the case of a rod-shaped molecule such as CS$_2$ is highly restricted by the interaction of neighboring molecules.\textsuperscript{10} The rotational motion of each molecule is affected by the field of the induced dipoles of the surrounding molecules.\textsuperscript{11} In addition, collision-induced anisotropy and, in particular, electron cloud overlap in liquids play an important role in determining the magnitude of the nonlinearity.\textsuperscript{11,12}

The reorientational Kerr effect in CS$_2$ was used to switch a visible light beam with an infrared light pulse.\textsuperscript{12-15} The measured value of the nonlinear refractive index caused by the reorientation of the molecules of CS$_2$ under picosecond excitation is of the order of $10^{-11}$ esu.\textsuperscript{1,2,12}

In the presence of a nonuniform electric field in any dielectric medium, the induced dipoles experience a force proportional to the gradient of the electric field strength. Since the induced dipoles are proportional to the electric field, the net force on each molecule is proportional to the gradient of the square of the field. This nonlinearity, which can give rise to self-focusing, is referred to as electrostriction.\textsuperscript{18,19} A microscopic treatment\textsuperscript{20,21} shows that the electrostrictive effect is influenced by the force experienced by the dipoles in the field of other dipoles and
the external field as well as the collisions, which do not add to zero because of the field induced inhomogeneity. On the edge of the beam where the gradient of the field is maximum the force on the individual molecules is also maximum. This leads to an increase of the density of the dielectric. A positive change of refractive index follows the change in the macroscopic density of the material. In general, because of the inertia, the electrostriction cannot have as fast a response time as the Kerr effect. The response time of electrostriction is determined by \( t = \frac{r_0}{V} \); where \( r_0 \) is the characteristic size of the incident beam and \( V \) is the speed of sound in the material. Physically, this is the time required for the density wave which is generated by the electrostrictive force at the edge of the beam to reach the center of the beam, where it will have the maximum effect on the self-focusing process. For a 10 \( \mu \text{m} \) beam size in a liquid this time is of the order of 10 ns. For picosecond excitations the change of the refractive index never reaches its steady-state value. The role of electrostriction, therefore, is negligible in picosecond experiments.\(^1,2,12\) For these experimental conditions the characteristic response time of the electrostriction is less than 30 ns. This compared to 270 ns pulse lengths used in these experiments shows that the contribution of the electrostriction to the change of refractive index cannot be ignored. Increasing the spot
size by a factor of two, as was the case in the photoacoustic method, increases the response time by a factor of two. Complete theoretical treatment of the interaction of light and the pressure wave in solids and liquids is performed respectively by Kerr\textsuperscript{22} and Lai.\textsuperscript{23} Their approximate and numerical solutions show that if the pulse duration is much longer than the characteristic response time of electrostriction, having all other parameters fixed, the effect is not sensitive to the details of the beam shape and beam size. Since the fundamental force responsible for the electrostriction depends on the gradient of intensity, the refractive index changes are independent of polarization of the incident beam. Electrostriction, therefore, produces an isotropic refractive index change and does not contribute to the induced birefringence. For this reason, in experiments where a weak beam probes the induced birefringence from a strong beam\textsuperscript{12,15} the contribution of the electrostriction is left out of the measurements. However, in all of the three experimental methods used in this work the contributions of the electrostriction and the Kerr effect to the change of refractive index cannot be separated.

The reorientational Kerr effect and the electrostriction have been considered as the two candidate nonlinearities which can lead to self-focusing in solids and liquids.\textsuperscript{24,25,30} The relative importance of these two
mechanisms in the self-focusing of pulse laser beams has been discussed.\textsuperscript{26,27} Shen\textsuperscript{28} has derived the electrostrictive coefficients and optical Kerr constants for some liquids. In the presence of an optical field, change of density $\Delta \rho$ obeys the driven acoustic wave equation

$$
[-\nabla^2 + (1/V^2) \partial^2 / \partial t^2 - (21/V^2) \partial / \partial t] \Delta \rho = -(1/8\pi) \gamma / V^2 (V^2 E^2)
$$

where $V=1.5 \times 10^5$ cm/s is the acoustic velocity in CS\textsubscript{2}, $\Gamma$ is the acoustic damping, and

$$\gamma = 2n_0\rho_0(\partial n/\partial \rho) = (n_0^2 - 1)(n_0^2 + 2)/3$$

is the electrostrictive coupling parameter. Shen\textsuperscript{28} solved the acoustic wave equation neglecting $\nabla^2 (\Delta \rho)$, which is valid for $r << r_0$ and assuming a Gaussian beam shape with $H W e^{-1} M$ of $r_0$ and a triangular pulse shape with FWHM of $t_p$. The ratio of the change of the refractive index due to electrostriction over that due to the orientational Kerr effect at the peak of the pulse is given by

$$
(\Delta n)_\rho / (\Delta n)_\alpha = (K_\rho/K_\alpha) (V^2/r_0^2) (1-r_0^2/r_0^2) t_p^2.
$$

$r$ is the radial coordinate, and $K_\rho$ is the electrostrictive coefficient and is calculated\textsuperscript{28} for CS\textsubscript{2} to be $2.53 \times 10^{-7}$. $K_\alpha$ is the optical Kerr constant and its values for several liquids and solids are calculated by Raman and Krishnan.\textsuperscript{29} For liquid CS\textsubscript{2}, $K_\alpha$ is $3.26 \times 10^{-7}$. With our 27 \mu m spot size and the 270 ns long pulses, this ratio on the axis of the beam ($r = 0$) will be approximately 130. In order to have a
value of one for this ratio, pulses of the order of 10 to 20 ns long should be used. It should be pointed out that neglecting $\nabla^2(\Delta \rho)$ is not a good approximation for very small beam sizes, and in solving the wave equation the component of the gradient of the electric field in the direction of the propagation is also neglected. Nevertheless this large ratio indicates that under the experimental conditions, the electrostriction is playing an important role and should be considered more seriously as the dominant mechanism responsible for the observed self-focusing.

In search of solid experimental evidence for the role of electrostriction in these experiments, I considered the temperature dependence and the polarization dependence of this nonlinearity as opposed to the Kerr effect. Electrostrictive coefficients have a strong positive temperature dependence,\textsuperscript{28,30} while the optical Kerr constants have a weaker negative temperature dependence.\textsuperscript{12,28,30-32} Physically, for electrostriction, this is because of the variation of the speed of sound and compressibility of the material, and, for optical Kerr effect, because of the increase of thermal random motion of the molecules with the increase of temperature. The critical power for self-focusing was measured at a higher temperature, using the photoacoustic method. Figure 4.1 shows the results of the measurement of the photoacoustic signal versus incident power at 10.55 $\mu$m for two
Figure 4.1—Photoacoustic signal versus incident power at 10.55 μm for temperatures of 22°C and 50°C.
different temperatures of 295 and 323 degrees Kelvin. At room temperature (22 degrees Celsius) the threshold power for self-focusing is $78 \pm 16$, which is in agreement with the previous measurement of $80 \pm 16$, in Chapter III. It is apparent that the threshold power for self-focusing decreases to the value of $71 \pm 15$ by increasing the temperature to 50 degree Celsius. This is only a 10% increase in absolute temperature and makes a 10% increase in the nonlinear refractive index. In agreement with our estimated result, this measurement shows that the electrostriction is the dominating nonlinearity responsible for the observed self focusing. However, this measurement does not rule out the presence of a smaller reorientational effect.

In order to further reinforce this point we consider the polarization dependence of electrostriction and the reorientational Kerr effect. As was pointed out the magnitude of the electrostrictive force is proportional to the gradient of intensity. Electrostriction, therefore, gives rise to an isotropic refractive index change independent of polarization of the incident electric field. However, the reorientational Kerr effect has a strong polarization dependence. Both linearly polarized plane waves and circularly polarized plane waves are solutions of the Maxwell equations with a nonlinear polarizability proportional to the field cubed. If the nonlinearity arises from the reorientation of the
molecules, the nonlinear refractive index experienced by linearly polarized light is different from that experienced by circularly polarized light. Calculated values of the intensity-induced changes of the refractive index are four times as great for linearly as for circularly polarized waves. This should be reflected in the critical power for self-focusing which can be measured with the photoacoustic method. Figure 4.2 shows the results of the photoacoustic measurements of the critical power for self-focusing for linearly and circularly polarized beams at 10.6 μm. The method of generation of the circular polarization was described in Chapter II. Figure 4.2 shows that the threshold power for self-focusing of linearly polarized beam is 154 ± 31 kW, and that for circular polarized light is 163 ± 33 kW. Taking the linear absorption at 10.6 μm into account and using the method described in Chapter III, we extract values of \( n_2 \) for the two polarizations. For the linearly polarized incident field we have \( n_{2L} = (3.29 \pm 0.66) \times 10^{-10} \) esu, and for the circularly polarized field we have \( n_{2C} = (3.12 \pm 0.62) \times 10^{-10} \) esu. These results show that the reorientational Kerr effect is but a small perturbation to the electrostriction in these experiments. The difference of \( n_2 \) for the two polarizations is only six per cent, which is less than the error bars of the experiment. For all practical purposes the two values of \( n_2 \) should be considered
Figure 4.2—Photoacoustic signal versus incident power at 10.6 μm for circular and linear polarizations of the incident beam.
equal. However, the uncertainty is in the measurement of absolute power of the beam. We may therefore consider the relative difference of two threshold powers to be real, and caused by the contribution of the orientational Kerr effect. From the scatter of data at the onset of nonlinearity shown in figure 4.2, we can estimate a relative error bar of 30 percent. The two values of \( n_{2l} \) and \( n_{2c} \) can be used to estimate the magnitude of the contribution of Kerr effect in these experiments. It was pointed out that for the orientational Kerr effect, \( n_2 \) for linearly polarized light should be four times greater than \( n_2 \) for circularly polarized light. Previous measurements of the nonlinear refractive index for linearly polarized and circularly polarized beams, however, show only a factor of two difference.\(^1,2,33\) These were under such experimental conditions that the electrostriction could be neglected (picosecond excitation) and the orientational Kerr effect was considered to be the dominant mechanism. Close\(^33\) attributes this lower ratio to the instabilities associated with the propagation of the circularly polarized beams in a cubic nonlinear medium. A linearly polarized light emerges linearly polarized from the sample but a circularly polarized wave is largely depolarized as soon as the threshold for self-focusing is reached.\(^33\) The disagreement between the theoretical value of four and the experimental value of two for the ratio of \( n_2 \) for linearly polarized over
n\textsubscript{2} for circularly polarized light is yet to be resolved. However, for the purpose of estimating the value of the nonlinear refractive index of CS\textsubscript{2} contributed by the Kerr effect in this experiment we use the more conservative value of two for this ratio. We can write

\[ n_{2\alpha l} = 2n_{2\alpha c} \]

where \( n_{2\alpha l} \) is the contribution of Kerr effect to the nonlinear refractive index for linearly polarized incident field and \( n_{2\alpha c} \) is that for circularly polarized incident beam. The nonlinear refractive indices measured from figure 4.2 have the two contributions of electrostriction and Kerr effect. Therefore, we can write for the linearly polarized beam

\[ n_{21} = n_{2\alpha l} + n_{2\rho} \]

and for the circularly polarized beam

\[ n_{2c} = n_{2\alpha c} + n_{2\rho} \]

Here, the subscript of 1 is for linear polarization, c is for circular polarization, \( \alpha \) is for reorientational Kerr effect, and \( \rho \) is for the electrostriction. Of course \( n_{2\rho} \) is the same for both polarizations. Using the measured values of \( n_{21} \) and \( n_{2c} \) we find \( n_{2\alpha l} = (3.4 \pm 1.9) \times 10^{-11} \) esu and \( n_{2\rho} = (2.95 \pm 0.8) \times 10^{-10} \) esu. It is apparent that the contribution of electrostriction is greater than Kerr effect approximately by an order of magnitude. The value of \( n_{2\alpha l} \) is also comparable to the value of \( 1.3 \times 10^{-11} \) esu measured in
CS$_2$ under picosecond excitation in the visible,\textsuperscript{1,2,12} which is only caused by the reorientational Kerr effect.

In Chapter III it was mentioned that the only other measurements of $n_2$ of CS$_2$ in the infrared region was reported by Owen\textsuperscript{15} and Jungling\textsuperscript{34} and that their results disagree by three orders of magnitude. Owen reported a value of $1.1 \times 10^{-11}$ esu, which is in remarkable agreement with $n_2$ reported for visible and near infrared. It is clearly evident that Owen has only measured the orientational contribution. He used a mode-locked TEA laser producing 250 ns long pulse train of 1 ns wide pulses. He then probed the birefringence in liquid CS$_2$ induced by the 10.6 $\mu$m radiation, using a helium neon laser with the CS$_2$ cell placed between cross polarizers. Although individual pulses in the mode-locked pulse were too short to be affected by the striction pressure created by themselves, the change of density created by one pulse could affect other pulses coming later in the 250-ns-long train of pulses. As it was pointed out earlier, because electrostriction does not induce any birefringence in the material, his experiment was sensitive only to the Kerr effect. On the other hand Jungling used a Q-switched CO$_2$ laser with smooth 200-ns-long pulses. By observing the beam shape transmitted through the CS$_2$ cell he estimated the value of the critical power for self-focusing. In this method the contributions of electrostriction and Kerr effect are mixed.
and cannot be distinguished. Jungling estimated a value of 5 kW for the critical power for self-focusing at 10.6 μm, which results in a nonlinear refractive index larger than reported in this work by two orders of magnitude. Comparison of only two beam scans after two different length CS₂ cells and arbitrary estimation of the critical power for self-focusing is not a very accurate or sensitive method. Nevertheless, the order of magnitude estimation of Jungling and the results of Owen confirm that electrostriction is playing an important role in the observed nonlinearity in this work.

**Stimulated Brillouin Scattering**

A discussion of intensity induced pressure waves generated because of electrostriction was introduced and the acoustic wave equation describing the density variation in the material was given in the previous section. Stimulated Brillouin scattering results when light waves couple to the acoustic wave.\(^{35}\) Two optical waves whose frequencies differ by \(\nu_s\) can drive an acoustic wave of this frequency. Similarly, a pressure wave of frequency \(\nu_s\) couples to an electromagnetic wave through the varying induced dipole moment density.\(^{36}\) The energy is transferred from the incident beam to the scattered beam. The forward Brillouin scattering does not occur because of its zero frequency shift. The back-scattered light is highly collimated and is
frequency down-shifted from the incident frequency $\nu_0$ by the value of $\Delta \nu = 2\nu_0 (V/c) \sin(\theta/2)$, which was first predicted by Brillouin. $^37$ $V$ and $c$ are the speed of sound and light, and $\theta$ is the angle of the incident and the scattered light (backward direction is $\theta = 180^\circ$). This frequency shift for some liquids, including $\text{CS}_2$, is measured to be of the order of $0.1 \text{ cm}^{-1}$, which corresponds to ultrasonic frequencies of the order of $10^9 \text{ Hz}$. $^38$ The emergence of the back-scattered Brillouin light and the build-up of the acoustic wave have a definite intensity threshold. This scattering process is completely described by the three coupled differential equations for the forward and backward traveling fields and the density variation of the material. Kroll $^39$ has worked out a steady state theory of the stimulated light scattering in a linearized approximation, ignoring saturation effects, for a number of different cases of material excitation. Tang $^40$ has given an analysis that takes saturation effects into account. It was shown that when the acoustic waves are heavily damped, such that the attenuation length is much smaller than the interaction region and the optical absorption length, the stimulated Brillouin scattering problem can be described by two coupled first-order nonlinear rate equations for the intensities of the laser and the scattered light. These steady-state theories were experimentally verified in $\text{CS}_2$ and other liquids, using pulses
from a Q-switched ruby laser, by Maier.\textsuperscript{41-42} He later studied transient Brillouin scattering in liquids experimentally.\textsuperscript{43, 44} Furthermore, Grob\textsuperscript{45} has confirmed results obtained by Maier\textsuperscript{41} in CS\textsubscript{2} by solving the exact coupled differential equations numerically under steady state conditions.

As it was shown in the previous section the steady state assumption is valid for the long pulses used in our experiment. Heavy damping of the acoustic waves is also valid, since attenuation distance for the acoustic waves in liquids is of the order of 20 $\mu$m, much smaller\textsuperscript{45} than the typical Rayleigh range in my experiment, which is to be considered as the interaction region for the Brillouin scattering. Assuming plane waves of infinite extent, ignoring linear absorption for the optical beams which would be small compared to the pump depletion due to Brillouin reflection, taking the laser and Brillouin frequencies to be approximately equal, and neglecting all other nonlinear processes such as self-focusing and Raman scattering, the evolution of Brillouin and laser intensity in $z$ (the propagation distance) will be described by\textsuperscript{40}

$$\frac{\partial I_B}{\partial z} = -gI_L(z)I_B(z)$$
$$\frac{\partial I_L}{\partial z} = -gI_L(z)I_B(z)$$

$I_L$, the laser intensity propagates in the $+z$ direction and $I_B$, the Brillouin intensity propagates in the $-z$ direction. $g$ is the gain factor and is given by\textsuperscript{40-42}. 

[Image]
\[ g = \left( \frac{k_L^2 \gamma^2}{2 \pi c n^3 \rho \delta \nu} \right) \times 10^7 \text{ cm/W}. \]

\( \gamma \), the electrostrictive coupling parameter, was given in the previous section; \( \rho \) is the density; \( V \) and \( \delta \nu \) the velocity and the linewidth of the acoustic wave, respectively. The numerical value of the gain factor depends on the literature values of the acoustic linewidth \( \delta \nu \) and the electrostrictive coupling parameter \( \gamma \). For \( \delta \nu \) we find the value of 63 MHz for \( \text{CS}_2 \) in references 43 and 46, and for \( \gamma \) we use the formula given in the previous section (which was shown to be a good approximation for \( \gamma \)) to find the value of 2.3. Using these parameters for the 10.6 \( \mu \text{m} \) laser radiation, the gain factor is calculated to be approximately 0.4 cm/GW. The threshold intensity for the buildup of the back-reflected light is defined by Tang to be

\[ I_{\text{th}} > \frac{2\alpha}{g}, \]

where \( \alpha = 0.21 \text{ cm}^{-1} \) is the optical absorption coefficient of \( \text{CS}_2 \) at 10.6 \( \mu \text{m} \). The value of \( I_{\text{th}} \) calculated for these experiments at 10.6 \( \mu \text{m} \) is 1.1 GW/cm².

The sharp rise of the acoustic signal in the photoacoustic method described in Chapter III could in principle be the result of the onset of the Brillouin scattering. However, in all the experiments neither back reflection nor pump depletion characteristic of Brillouin scattering was observed. The intensity available at the focus, before the onset of self-focusing, for all of the experiments, was less
than the calculated intensity threshold for the stimulated Brillouin scattering. The results obtained in this section, confirmed experimentally by the dependence of self-focusing threshold on power, rather than intensity, show that the stimulated Brillouin scattering could not have an active role in the measurements of the nonlinear refractive index of CS$_2$.

The Laser-Induced Thermal Effects

Thermal effects result from the nonlinear interaction of the laser radiation with the material, heated by absorption of the radiation itself. The energy absorbed from the laser beam changes the temperature, which in turn changes the density of the material. This gives rise to a variation of the refractive index through its dependence on temperature and density. The intensity distribution of the laser beam will then be affected by the changes in the refractive index. Unlike self-focusing, this process does not have a threshold. A small absorbed energy causes a small change in the refractive index. However, for sufficiently long times (msec to sec) a steady state is achieved that can be characterized by the form of heat transfer which balances the absorbed laser power. For most materials the rate of change of index of refraction with respect to temperature is negative, and thus the thermal distortion causes a decrease in the on-axis intensity as the beam propagates through the material. This is referred to as self-defocusing. The problem is described
by the same equation as self-focusing with the exception that $n_2$ is negative for thermal self-defocusing. The index of refraction of some materials, such as Germanium and water below 4 °C, increases with temperature, leading to thermal self-focusing. Also, in most semiconductors the nonlinearity arises from the temperature dependence of the band gap which can result in thermal self-focusing. However, these electronic thermal effects are virtually noninertial and have characteristic times of the order of picoseconds. For liquid CS$_2$ the rate of variation of the index of refraction with respect to temperature at constant pressure is $-80.5 \times 10^{-5} \text{C}^{-1}$ measured at 1 atm. and the rate of change of refractive index with respect to pressure at constant temperature is $66.81 \times 10^{-6}$ measured at 22 °C. Some of the early investigations of the thermal effects were concerned with the low-power CW visible lasers. Buildup and decay transients, mode changes, and relaxation oscillations, all with time constants of the order of 0.1 to 1 second, were observed when liquid cells were placed inside a He-Ne laser cavity. The phenomenon was demonstrated to be useful for measuring low optical absorption of liquids and construction of a power-limiting device. The effect of the thermal lens on the beam shape of a He-Ne laser propagating through an external CS$_2$ cell was photographed by Callen. Thermal effects can deteriorate the quality of beams
 propagating through the atmosphere. The effects of wind and convection were studied by Hull.\textsuperscript{55} A comprehensive review of the thermal effects under steady state and transient, collimated and focused beam, single pulse and pulse train is given by Smith.\textsuperscript{56} In addition the influence of molecular and aerosol absorption and relaxation, laser beam jitter, turbulence, convection and flow of the material is discussed. Liu\textsuperscript{57} showed that in general there is a small cooling effect because of the expansion of the material. Buser\textsuperscript{58} studied the transient thermal effect and showed that, neglecting viscosity and thermal conduction, the characteristic time required for the laser to cause a density change in the material equal to $\rho_0$ is

$$t_{cl} = \frac{3\pi r_0^2 \rho_0^2}{2\alpha P},$$

where $r_0$ is $H\omega e^{-2} M$ of intensity, $\rho_0$ is the density, $V$ is the speed of sound, $\alpha$ is the absorption coefficient, and $P$ is the power of the beam. Buser confirmed this relation with 7-ms pulses chopped from a CW $CO_2$ laser propagating through an absorbing gas cell. He found time constants for the buildup of the thermal lens of the order of 100 milliseconds. Using typical parameters for experimental conditions, this time is of the order of 10 $\mu$s. Considering thermal conduction and diffusion, the characteristic time for the establishment of the stationary temperature gradient is calculated\textsuperscript{34,51,59-61} to be
Here, \( k = 4 \times 10^{-4} \text{ cal/(cm.s.°K)} \) is the thermal conductivity, and \( C_p = 0.24 \text{ cal/(g.°K)} \) is the specific heat for CS\(_2\).\(^47\) In a pump-probe experiment the time required for the thermal lens to be established with an unfocused TEA CO\(_2\) laser was measured to be 200 ms.\(^61\) This time is of the same order of magnitude as the time constants measured by Gordon\(^51\) in the experiments with the He-Ne laser. For my experimental conditions, I calculated the characteristic time for decay of the thermal lens to be of the order of one millisecond. In an experiment these time constants were measured by probing the thermal lens with an He-Ne beam. The He-Ne beam was propagating in the same direction as the CO\(_2\) pulse and was focused by the same lens LI. A fast photodiode behind a pinhole and a glass plate (to block the CO\(_2\) pulse) monitored the on-axis intensity of the He-Ne. The He-Ne beam diverges after passing through the thermal lens created by the CO\(_2\) pulse. The photograph of the oscilloscope trace of the signal is shown in figure 4.3. The thermal lens realizes its maximum strength after 200 µs and decays in a much longer time, on the order of a few ms. Clearly the thermal effect, compared to the pulse length of the CO\(_2\) laser, is too slow to play a significant role during the propagation of the pulse. This was experimentally verified in Chapter III. Changing the focal length of the lens LI in the photoacoustic
Figure 4.3--The photodiode signal versus time displayed on the oscilloscope.
experiments had no effect on the critical power for self-focusing, whereas the temperature rise because of absorption in the focal volume was greater by a factor of four when the beam was focused by the smaller focal length lens. Because of the opposing roles of the thermal effect and self-focusing, a smaller threshold for self-focusing would be expected with the CO\textsubscript{2} beam focused by the smaller focal length lens. The results of the photoacoustic experiments, therefore, show that the negative contributions of the thermal effect to the self-focusing problem are negligible in our measurements. The role of thermal defocusing is directly linked to the linear absorption of the material. In the next section we will examine the effect of linear absorption on the self-focusing problem.

The Effect of Linear Absorption on Self-Focusing

In Chapter III the relation between the critical power and the threshold power for self-focusing was given with an empirical formula. In general the interaction of the linear absorption and self-focusing can be taken into account by including the linear absorption in the nonlinear wave equation which governs the propagation of the beam in the material. This is what was done in our numerical simulation. In extracting $n_2$ from the results of the photoacoustic measurements it was assumed that the energy of the beam
depletes through the linear absorption mechanism without any significant self-focusing until the beam has reached to the focal area. Dawes\textsuperscript{62} and Marburger\textsuperscript{63} with numerical computation showed that as long as the absorption length is longer than the self-focusing length, increasing absorption does not prevent the collapse of the beam to a singularity. This point was confirmed in our own numerical simulations. Increasing the absorption coefficient only moved the singular point to a longer distance, but it was not removed until the absorption length was reduced to be smaller than the self-focusing length. We will show in this section that although the threshold power for self-focusing increases with absorption, the critical power for self-focusing remains constant. This is because of the depletion of power before the focal area and is consistent with the picture presented in Chapter III. To my knowledge, the only other experimental study of the effect of absorption on self-focusing was done by Wang,\textsuperscript{64} using Q-switched ruby laser in CS\textsubscript{2}. The absorption coefficient was varied by dissolving Iodine in CS\textsubscript{2}. In his work the approximate theory developed by Kelley\textsuperscript{65} was used. The threshold power for self-focusing was taken as the threshold for stimulated Raman emission. His results showed, in disagreement with his theory, that the critical power for self-focusing was not constant. Wang attributed this disagreement to the over-simplified theory used to
calculate the critical powers. It is interesting to note that their results showed that the critical power for self-focusing was greater for smaller linear absorption which cannot be explained by the influence of thermal defocusing.

I studied the effect of linear absorption on the critical power for self-focusing experimentally. Following Wang, the linear absorption coefficient $\alpha$ was varied from 0.002 cm$^{-1}$ to 0.81 cm$^{-1}$ by dissolving a controlled amount of iodine in CS$_2$. The frequency-doubled pulses at 0.53 $\mu$m from a mode-locked YAG laser were used for this experiment. Because linear absorption of CS$_2$ in the infrared is large, the experiment was performed at 0.53 $\mu$m, where the absorption coefficient is very small. The linear absorption in the visible can be varied over a wide range. With 50 ps pulses of the YAG laser the dominating nonlinearity is the reorientational Kerr effect. Although the important experimental parameters such as wavelength, linear absorption, and pulsewidth, are different from these infrared experiments, the influence of linear absorption on self-focusing is the same in both experimental domains.

The threshold power for self-focusing was measured, using the power-limiting method. The method and its experimental setup are described in Chapter III. The single transverse mode beam of the size $r_0 = 1$ mm was focused with $L_1 = 7.5$ cm focal length lens in the center of the 2 cm long CS$_2$ cell.
The linear dependence of absorption coefficient on the concentration of iodine was verified with a spectrophotometer. The rate of change of linear absorption coefficient at 0.532 µm with respect to the concentration of iodine was measured to be $1.84 \times 10^3$ cm$^2$/gr. In order to make sure that there was no optical nonlinearity associated with the iodine, different concentrations of iodine in alcohol were tested in the same setup. For incident powers ten times greater than $P_c$ for CS$_2$ and concentrations of up to $4.34 \times 10^{-3}$ gr/cm$^3$, no nonlinear refraction, absorption or saturation was observed. Figure 4.4 shows the results of the power-limiting experiment for pure CS$_2$ and CS$_2$ with iodine concentration of $4.4 \times 10^{-4}$ gr/cm$^3$. The absorption coefficient corresponding to this concentration is 0.81 cm$^{-1}$, and pure CS$_2$ at 0.532 µm has an absorption of 0.002 cm$^{-1}$. The critical power for self-focusing for pure CS$_2$ from this experiment is seen to be 7.03 kW. This gives a nonlinear refractive index of $1.44 \times 10^{-11}$ esu, which is in good agreement with the previous measurements with picosecond excitation at this wavelength.$^{1,2,12}$ At the higher absorption of 0.81 cm$^{-1}$ the threshold power for self-focusing increases, by approximately a factor of three, to 19.2 kW. Taking the absorption into account, this corresponds to a critical power of 8.5 kW, which is only 20% different from that for the pure CS$_2$. Table IV shows the results of a set of experiments for some other
Figure 4.4—The signal on D3 versus incident power at 0.53 μm for pure CS₂ and 4.4 x 10⁻⁴ gr/cm³ of Iodine in CS₂. The focus is positioned in the center of the sample (l = 1 cm).
intermediate absorption coefficients at 0.53 μm with the focus at the center (l = 1 cm) of the CS₂ cell.

TABLE IV
THE RESULTS OF MEASUREMENTS OF THE THRESHOLD AND CRITICAL POWER FOR SELF FOCUSING VERSUS ABSORPTIONS FOR l = 1 cm

<table>
<thead>
<tr>
<th>C x 10⁵ gr/cm³</th>
<th>α cm⁻¹</th>
<th>P_t kW</th>
<th>P_c kW</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.002</td>
<td>7.04</td>
<td>7.03</td>
</tr>
<tr>
<td>4.4</td>
<td>0.082</td>
<td>7.52</td>
<td>6.93</td>
</tr>
<tr>
<td>8.8</td>
<td>0.162</td>
<td>8.0</td>
<td>6.81</td>
</tr>
<tr>
<td>13.2</td>
<td>0.243</td>
<td>8.32</td>
<td>6.55</td>
</tr>
<tr>
<td>17.6</td>
<td>0.323</td>
<td>8.8</td>
<td>6.47</td>
</tr>
<tr>
<td>26.4</td>
<td>0.484</td>
<td>10.56</td>
<td>6.54</td>
</tr>
<tr>
<td>44.0</td>
<td>0.806</td>
<td>19.2</td>
<td>8.47</td>
</tr>
</tbody>
</table>

The results presented in Table IV are plotted in figure 4.5. The average value of the critical power is 6.96 ± 0.65 kW. It is clear that although the threshold power for self-focusing P_t changed by a factor of four, the critical power remained constant to within 10 percent. In the next set of experiments, keeping all the conditions the same, the focus of the beam was moved toward the front surface of the cell (l = 1 mm). Since in this case the beam does not lose significant power before reaching the region of nonlinear interaction (the Rayleigh range), it was expected that the threshold power as well as critical power would be independent
Figure 4.5--The threshold ($P_t$) and critical ($P_c$) power for self-focusing versus linear absorption coefficient with the focus at the center of cell.
of absorption. The results of the power limiting experiments under this condition for pure CS\textsubscript{2} and $4.4 \times 10^{-4}$ gr/cm\textsuperscript{3} of iodine in CS\textsubscript{2} are shown in figure 4.6. It can be seen that the variation of the threshold power is less than 5%, with a large increase in linear absorption. Table V summarizes the results of the experiments for other intermediate absorption coefficients at $0.53 \mu m$ with the focus at nearly the front surface ($\ell = 1$ mm) of the CS\textsubscript{2} cell. The results presented in Table V are plotted in figure 4.7.

**TABLE V**

THE RESULTS OF MEASUREMENTS OF THE THRESHOLD AND CRITICAL POWER FOR SELF FOCUSING VERSUS ABSORPTION FOR $\ell = 1$ mm

<table>
<thead>
<tr>
<th>$C \times 10^5$ gr/cm\textsuperscript{3}</th>
<th>$\alpha$ cm\textsuperscript{-1}</th>
<th>$P_t$ kW</th>
<th>$P_c$ kW</th>
</tr>
</thead>
<tbody>
<tr>
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Figure 4.6--The signal on D3 versus incident power at 0.53 μm for pure CS₂ and 4.4 x 10⁻⁴ gr/cm³ of Iodine in CS₂. The focus is positioned at \( l = 1 \) mm inside the sample.
Figure 4.7--The threshold ($P_t$) and the critical ($P_c$) power for self-focusing versus linear absorption coefficient with the focus positioned at $\ell = 1$ mm inside the sample.
In this experiment the average value of $P_t$ is $7.25 \pm 0.19$ kW and that for $P_c$ is $6.98 \pm 0.14$ kW; these are virtually constant. These results show that the critical power for self-focusing is independent of absorption and that the method of extraction of $n_2$ described in Chapter III is valid.

Conclusion

In this chapter we have examined in some detail the relevant physical mechanisms responsible for the observed nonlinearity in these experiments. The reorientational Kerr effect and the electrostriction were shown to be responsible for self-focusing in CS$_2$, with electrostriction having the dominant role. It was shown that this is because the acoustic transit time is much smaller than the pulse duration, so that a steady state is achieved for the electrostriction pressure. This point was further reinforced experimentally by the temperature and polarization dependent measurements. A ten per cent decrease in the critical power was observed as a result of approximately ten per cent increase of the absolute temperature of the CS$_2$. This increase in the strength of the nonlinearity is expected if electrostriction is dominant, whereas the opposite is expected if the Kerr effect is the dominant nonlinearity. The relative magnitude of the electrostrictive and Kerr contributions to the nonlinear refractive index was estimated by measuring the critical power for self-focusing with linear
and circular polarizations. These results show that in these experiments the electrostriction is stronger than the Kerr effect by one order of magnitude. The stimulated Brillouin scattering was not observed in these experiments. The intensity threshold for this process was calculated based on a steady state theory. The intensity at the focus in all the experiments was less than this value. In order to observe stimulated Brillouin scattering, higher intensities and longer interaction lengths are required. These calculations, as well as the experimental evidence of the power dependence of the threshold powers for the onset of the nonlinearity, show that the sudden increase in the acoustic signal is not initiated by stimulated Brillouin scattering. The question of the thermal effect was also considered. Time constants involved in this process were estimated. Neglecting viscosity and thermal conduction, the response time was calculated $^{58}$ to be approximately 10 μs. Taking thermal conduction into account, this time was of the order of milliseconds. $^{51}$ Probing of the thermal lens created by CO$_2$ pulse with a He-Ne laser confirmed these long response times. Such slow responses rule out the influence of the thermal effect during CO$_2$ laser pulses. This point is reaffirmed by considering the effect of the linear absorption on self-focused through the direct link between the absorption and thermal effect. What was seen in numerical simulations here
and by others\textsuperscript{62,63} was confirmed in an experiment where the linear absorption was varied by dissolving iodine in CS\textsubscript{2}. It was shown that the critical power for self focusing, and therefore the nonlinear refractive index, are independent of linear absorption. This in effect was shown in a different way in Chapter III by using the beam distortion measurement. It was demonstrated that by including the linear absorption into numerical solutions of the wave equation, a good fit was obtained for different positions of the focus in the samples all having the same n\textsubscript{2}. In this experiment the absorbed power before the focus was changed by varying the distance of the focal point from the entrance face of the sample rather than varying the absorption coefficient. The variation of the phase front of the beam was taken into account in the numerical computations.
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CHAPTER V

DISCUSSION AND SUMMARY

The microscopic physical mechanisms responsible for the observed nonlinearity are considered in Chapter IV. The roles of electrostriction, Kerr effect, stimulated Brillouin scattering, thermal effect, and the influence of absorption on nonlinear propagation are discussed in some detail. Some of the basic assumptions used in the interpretation of the experimental results are verified experimentally. The theoretical calculations are also consistent with these assumptions. It is shown that the increase of the nonlinear refractive index measured in these experiments compared to that reported in the visible\textsuperscript{1-3} can in large part be explained by the electrostrictive contributions. The observed variation of the nonlinear refractive index in the infrared region described in Chapter III will be discussed in this chapter. Up to this point we have been concerned with showing that the observed dispersion is real. This point is specifically emphasized in the consideration of the effect of linear absorption. It is concluded that the dispersion in $n_2$ is not an artifact of the way in which linear absorption is taken into account in reducing the experimental results while calculating $n_2$. In section one of this chapter we will
consider the possible origins of the dispersion. Some suggestions for future experiments are also given in this section. In the second and final section of this chapter a summary will be presented.

**Dispersion**

We first examine the electrostriction, since it is the dominating nonlinearity in our experiments. It has already been pointed out in Chapter IV that the magnitude and the temporal response of the electrostriction depend on the thermodynamic properties of the material. These include the isothermal compressibility, temperature, density, specific heat, the speed of sound, and the attenuation coefficient for the sound wave. As introduced in Chapter IV, the frequency of the sound wave generated by the interaction of the light with the material, depends directly on the frequency of the incident light beam. This is the same frequency shift for the stimulated Brillouin scattering and is of the order of $10^8$ Hz for the infrared excitation. The propagation behavior of these ultrasonic waves, therefore, has a direct bearing on the nonlinear interaction of the material with incident light through electrostriction. This is specially true in the case of long pulses (comparable to the time it takes for the sound wave to travel through an absorption length) such as the ones used in my experiments. The theory of the propagation of the ultrasonic waves through liquids is discussed by Andreae and
Lamb. The propagation of sound wave through liquids is described in terms of the absorption coefficient and the velocity. Postulating that the frequency variation arises only because of the internal degrees of freedom, the authors derive these acoustic quantities in terms of the thermodynamic parameters which describe specific molecular processes. The vibrational, translational, and rotational degrees of freedom, as well as chemical reactions in which molecules exchange their constituent atoms, are considered to be the different relaxation mechanisms responsible for the dispersion of the velocity and absorption of the ultrasonic waves. This frequency dependence is also reflected in the isothermal compressibility and the specific heat, which is the sum of components due to vibrational, rotational and other internal causes both thermal and structural. However, it is reasonable to assume that at any single frequency only one of these processes is contributing to the specific heat. The experimental evidence of the dispersion of the absorption of ultrasonic waves at room temperature is presented by Andreae and Lamb for some liquids. Their results for CS₂ show that the absorption coefficient for the ultrasonic waves increases by a factor of 6 when the frequency changes from 20 to 70 MHz. The expected relaxation times for the excitation of the rotational energy levels are too small to cause an ultrasonic dispersion in the neighborhood of 70 MHz, as
indicated in their results. Therefore, they consider the normal vibrational modes of the CS$_2$ to be the relaxation mechanism responsible for the measured dispersion. Their subsequent theoretical calculations of the specific heats and the associated velocity dispersion for the three different vibrational normal modes of CS$_2$ lead to the conclusion that the degenerate bending vibration ($\nu_2 = 397$ cm$^{-1}$ of Chapter III) is responsible for the observed behavior. In a separate study Andreae et al. extend the measurements of the dispersion of ultrasonic waves in liquid CS$_2$ to higher acoustic frequencies as well as different temperatures. The measurements are performed from 2 to 200 MHz and at temperatures of 25°C and -63°C. Their results indicate that the absorption coefficient for the ultrasonic wave in CS$_2$ at 25°C continues to increase with frequency to a value of 272 cm$^{-1}$ at the frequency of 189 MHz. The results of this extended study show that the observed behavior is not due to the time of transfer of energy to the degenerate bending vibration only, but rather to a relaxation of the total vibrational specific heat. The sum of the calculated values of the specific heat contributed by the three vibrational modes is used to calculate the absorption coefficients, and agreement of better than two per cent is obtained throughout the frequency range and for the two temperatures of 25°C and -63°C. The frequency of the ultrasonic waves generated by the
propagation of the CO₂ laser pulse in my experiments due to electrostriction varies from 300 to 400 MHz as the laser wavelength is tuned from 11 to 9 μm. This frequency range is close to the range of the reported anomalous dispersion of absorption and velocity of the ultrasonic waves in liquid CS₂. As it is pointed out, the electrostrictive properties of the material are sensitive to the thermodynamic parameters mentioned above. It is therefore reasonable to expect that the frequency dependence of the velocity and absorption coefficient of the sound wave, the specific heat, and the isothermal compressibility result in a dispersion of the electrostrictive properties of the material. As pointed out by Cummins, this is particularly important for CS₂ in the frequency range mentioned. Cummins studied the effect of the dispersion of the electrostriction on the process of stimulated Brillouin scattering. By including the velocity dispersion of the sound wave in the theory, he was able to resolve the existing disagreement between the calculated and measured values of the ratio of the Rayleigh and the Brillouin intensities. This is particularly evident for carbon disulfide since the theory without dispersion is clearly in disagreement with the experiment, whereas the inclusion of dispersion leads to agreement between theory and experiment. Tiganov used measurements of the intensity, position, and width of the Brillouin components to study the
dispersion properties of the ultrasonic waves in liquids. He points to the negative dispersion of water and acetone at room temperature. Relaxation processes of the type mentioned by Andreae\(^4\text{"}^{6}\) lead to a positive dispersion (velocity of sound increases with frequency). The sign of dispersion depends strongly on the intermolecular interaction. If a model of hard spheres which interact elastically is used, we have positive dispersion. If the molecules are bound by elastic forces, we can have negative dispersion. In all the experimental works mentioned above,\(^7\text{"}^{9}\) the ultrasonic waves are generated by the interaction of radiation of an He-Ne laser with liquids, including CS\(_2\). In these experiments the frequencies are of the order of \(10^9\) to \(10^{10}\) Hz. Their results indicate that the effect of dispersion of sound waves on the interaction of light with the material is significant. Although stimulated Brillouin scattering was not observed in my experiments, the physical mechanism responsible for the generation and absorption of the ultrasonic waves is the same as that in the Brillouin process. As pointed out by Andreae\(^5\text{"}^{6}\) the origin of dispersion of sound in the case of CS\(_2\) in the neighborhood of 200 MHz is the relaxation of the total vibrational specific heat of the molecule. This is very close to the sound frequencies present in our experiments. The situation is further complicated because the infrared radiation is close to resonance with the vibrational
modes of CS$_2$. Therefore a quantitative thermodynamic analysis of the magnitude of electrostrictive dispersion of the type performed by Andrea$^{4-6}$ is not possible because of the presence of the radiation in this work. However, the stimulated Brillouin scattering can be utilized to study the dispersive characteristics of electrostriction. For such studies, higher intensities and longer interaction lengths than the ones used in these experiments are called for. Measurements of frequency shift, width, and intensity of the scattered light at some angle must be performed. The temperature must be controlled since the thermodynamic parameters generally depend on temperature, and their values, needed for the calculation and comparison, are usually tabulated at a few temperatures in the literature. The samples must be dust-free, since their scattered light contributes to the central unshifted line in the spectrum, increasing the error of measurement. The scattering angle should be known accurately because one per cent error in the scattering angle causes one per cent error in the frequency shift. Care must be exercised in avoiding self-focusing, which has a definite power threshold.

It was pointed out in Chapter IV that the optical Kerr effect arises from the alignment of the molecules with the electric field of the light beam. The induced birefringence causes a phase difference between the two polarization
components of a probe beam traveling through the same region of the material. This is referred to as the optical Kerr effect, as opposed to the dc electro-optical Kerr effect in which the birefringence is induced by a static electric field. The first observation of variation of the magnitude of the birefringence in the dc Kerr effect with the frequency of the incident radiation was made by Kerr.\textsuperscript{10} Although the classical Drude-Lorentz\textsuperscript{11} theory was remarkably successful in explaining the phenomenon of ordinary dispersion and absorption, it was notably unsuccessful when directly applied to the electro-optic Kerr effect. Havelock\textsuperscript{12} was among the first to formulate a hypothesis which explained the observed dispersion. He considered the effect as due to the rearrangement in grouping of an assemblage of similar particles. His result, which is known as Havelock's law, predicts that the magnitude of the electro-optic Kerr effect depends inversely on wavelength of the incident light. Havelock's prediction is experimentally verified repeatedly in the visible\textsuperscript{13} and near infrared\textsuperscript{14} for liquid CS\textsubscript{2}. The theory of Kerr effect has since been completely revised.\textsuperscript{15-18} The induced anisotropy is now known to be due to the reorientation of the molecules. Nevertheless, Havelock's law is in qualitative agreement with the experimental results in non-absorbing regions of the spectrum.\textsuperscript{19} Early experimental work indicated a deviation of dispersive behavior of liquids from
Havelock's prediction, close to electronic and vibrational resonances of molecules.\textsuperscript{20} Later the theory of dispersion of the Kerr effect near absorption bands was developed by Buckingham.\textsuperscript{21,22} In his work the quantum theory of the Kerr effect is developed for gases and applied to the vibration-rotation spectral region of diatomic and symmetric top molecules. His results show that unlike the transparent regions, where the reorientation of the anisotropic molecules is the major contributor to the Kerr effect, in absorption bands distortion of the molecular structure by the strong electric field is the dominant effect. Buckingham\textsuperscript{22} showed that, close to an absorption line, the rapidly varying Kerr effect may be six orders of magnitude larger than that for the transparent region of the spectrum. Charney\textsuperscript{20} studied the dispersion of the electro-optic Kerr effect in nitrobenzene from 1.5 to 5.6 $\mu$m, which is the region of vibrational resonance for this molecule. His experimental results show that the anomalies of the refractive index in the neighborhood of resonant absorptions are accompanied by anomalous dispersion in the electro-optic Kerr effect. A theoretical analysis of the effect of the molecular structure on the Kerr effect, based on a forced-damped oscillator model, demonstrated that the form of dispersion in the neighborhood of a vibrational resonant absorption is dependent on the orientation of the dipole moment developed by the
normal mode corresponding to that absorption. Buckingham\textsuperscript{21} suggested that this effect can be used for assigning vibrational and electronic absorption bands. Recently the theoretical and experimental works of Bishop\textsuperscript{23} and Elliott\textsuperscript{24} show that the vibrational contribution to the third order polarizabilities is significant in absorbing regions of the spectrum. It has been shown\textsuperscript{25,26} that when a rapidly oscillating electric field is applied to a Kerr cell, the magnitude of induced birefringence changes dramatically with the frequency of the applied field. This effect in extremely viscous liquids has been observed\textsuperscript{27} in the frequency range of 60 to $10^7$ Hz, in qualitative agreement with Debye's theory.\textsuperscript{15}

The fundamental physical mechanisms present in the optical and dc Kerr effect are of the same nature, but the theories mentioned in the previous paragraph cannot be directly applied to the dispersion of the optical Kerr effect. This is because in the electro-optic Kerr effect the birefringence is induced and dispersion is experienced by different fields, whereas, in the optical Kerr effect, the reorientation of the molecules is caused by the same light beam. A discussion of this phenomenon is given in Chapter IV. Buckingham,\textsuperscript{28} the first to predict that birefringence can be induced by an intense light beam, indicated the difficulties associated with the observation of such phenomenon because of the lack of intense light sources.
With the availability of intense laser sources the optical Kerr effect has been observed by many researchers and many aspects of this effect have been treated. The dispersion of the third-order susceptibilities arising from fast electronic effects has been studied in the atomic systems. However, there is no theoretical treatment of the dispersion of the reorientational optical Kerr effect available in the literature, and experimental observations are scarce. The data available for CS\textsubscript{2} show no variation in the Kerr effect in the range of 0.5 to 1 μm. However, in the infrared, which for CS\textsubscript{2} is the region of dispersion of the linear index\textsuperscript{30} and resonance absorption, a large dispersion is expected. My estimated value of nonlinear refractive index associated with the Kerr effect in the infrared is larger than $n_2$ in the visible by a factor of three. In the absence of a complete theory of dispersion for the optical Kerr effect in the neighborhood of vibrational resonances, it is difficult from these results to make any conclusive remarks about the origin of the dispersion of the Kerr effect. This is particularly true in the presence of ultrasonic waves in my experiment, because, as is shown by the thermodynamic theory of Keyes,\textsuperscript{31} it is possible for the reorientational motion of the molecules to couple with both the transverse and longitudinal sound waves propagating in the liquid. The evidences of this coupling have been observed in the depolarized light
scattering spectrum. It is therefore essential for the study of the dispersion of optical Kerr effect in the neighborhood of the vibrational resonances to eliminate the effects of electrostriction. This can be achieved by using short pulses of the order of picoseconds. For such short pulses ultrasound waves are too slow to have any effect on the experimental results. With these short pulses the photoacoustic method can be used at high repetition rates to enhance the signal-to-noise ratio without the influence of slow processes such as thermal or electrostriction effects. Care must be exercised to avoid the stimulated scatterings, which have a definite intensity threshold. This experiment is in the planning stages.

The application of a strong electric field in a material can change the absorption spectrum. This effect has been known (Stark effect) for many years. In molecules the effect can arise because of the distortions of the electron cloud or the molecular structure. This effect has been used for the determination of excited-state dipole moments of some liquids.\(^{32,33}\) Dows,\(^{34}\) in an experimental study of \(\text{CS}_2\) in its \(R\) and \(S\) vibrational band, concludes that the dc field-induced spectrum arises because the \(\text{CS}_2\) is bent in its excited state and possesses a dipole moment. He observed no field-induced spectrum at shorter wavelengths (V-band region), suggesting that the molecule is not bent at the corresponding excited state.
The induced changes in absorption are of the order of one percent for an electric field strength of 30 kV/cm. Changes in the absorption can give rise to variations in the index of refraction. The connection between absorption spectrum and dispersion of refractive index is given by the Kramers-Kronig relation. This is of very general validity, and results from mere assumption of the causal connection between the polarization and the electric field. The electric fields typically present at the focus in my experiments are of the order of $10^7$ V/cm. The absorption coefficient of CS$_2$ at 10.6 μm was given in Chapter III to be 0.21 cm$^{-1}$. As pointed out, this measurement was performed at very low intensities using a spectrophotometer. The result of measurement of absorption using intense CO$_2$ laser pulses is approximately 0.4 cm$^{-1}$. It is therefore reasonable to expect intensity-induced absorption to make a significant contribution to the observed dispersion.

**Summary and Conclusion**

The nonlinear refractive index, $n_2$, of liquid carbon disulfide in the wavelength range of 9 to 11 μm were measured. For these measurements, 270-ns-long pulses of a tuneable hybrid TEA CO$_2$ laser were used. The photoacoustic technique was developed as a sensitive method for measurement of the onset of nonlinearity. The role of self-focusing as the dominant nonlinearity responsible for the abrupt increase
in the photoacoustic signal was established by varying the focal intensity while keeping the power constant. The onset of nonlinearity occurs at a certain threshold power for input intensities different by a factor of four which is the expected behavior if self-focusing is dominant. This indicates that the observed material breakdown is initiated by self-focusing. At the same time it rules out a significant contribution from intensity-dependent nonlinearities such as thermal variation of the refractive index and stimulated Brillouin scattering. Since the presence of these two nonlinearities could greatly alter the interpretation of the experimental results, they are discussed at some length.

In the photoacoustic method the nonlinear increase of the acoustic signal is seen as the result of collapse of the beam due to self-focusing. This interpretation is valid if the stimulated Brillouin scattering is not present before self-focusing turns on. In all the measurements, the back-scattered light from the CS$_2$ sample was monitored. No indication of an increase in the back-reflected light or pump depletion characteristic of stimulated Brillouin scattering was present. The focal intensity available before the onset of self-focusing in all experiments was smaller than the calculated value of the threshold intensity for this nonlinearity. The steady-state self-focusing as a model for the observed nonlinearity and the accuracy of the results
obtained by the photoacoustic method were verified by two other methods.

The nonlinear refractive indices obtained by the beam distortion measurement and power limiting techniques are in remarkable agreement with the results of the photoacoustic method. The validity of the method of extraction of \( n_2 \) from the measurements of the threshold power and the way in which the effect of linear absorption is taken into account has been experimentally studied. It has been verified that the important parameter is the power at the focus, rather than incident power, which determines the on-set of self-focusing. This is because the nonlinear interaction occurs mainly in the focal region where the intensity is greatest. It is shown that increasing linear absorption increases the threshold power but does not change the critical power for self-focusing. The critical power for self-focusing in the spectral range of the CO\(_2\) laser was calculated from the measured threshold power. Measurements of the critical power show a variation as large as a factor of four, resulting in a proportional change in the nonlinear refractive index in this spectral region. The results of these measurements indicate a maximum nonlinear refractive index of \((11.65 \pm 2) \times 10^{-10}\) esu at 10.27 \(\mu\)m and a minimum of \((3.13 \pm 0.6) \times 10^{-10}\) esu at 10.6 \(\mu\)m. These values are to be compared with \(1.3 \times 10^{-11}\) esu measured in the visible and near-infrared.\(^1\)-\(^3\)
In consideration of the effect of linear absorption discussed in Chapter IV, the critical power for self-focusing in solutions of iodine in CS$_2$ at 0.53 $\mu$m is measured to be 7 kW, using the power limiting technique. The nonlinear refractive index calculated from this measurement is $1.45 \times 10^{-11}$ esu, independent of the concentration of iodine (linear absorption). This is in good agreement with the values reported in the literature.$^{1-3}$

The physical origin of the nonlinearity in CS$_2$ was considered. Theoretical consideration of the electrostriction and the reorientational Kerr effect indicates that the former is the dominating microscopic process in these experiments. The temperature-dependent measurement confirms this result. At the same time, measurement of the nonlinear refractive index with the circular polarized light allows us to estimate the contribution of the electrostriction and the Kerr effect. The estimated value of the reorientational $n_2$ from this measurement is $(3.4 \pm 1.9) \times 10^{-11}$ esu, which is a factor of three greater than that reported in the visible and near-infrared under picosecond excitation,$^{1-3}$ while the estimated ratio of the electrostrictive $n_2$ to the $n_2$ contributed by the Kerr effect is 8.7. These results indicate that the larger value of $n_2$ in these experiments, compared to the results obtained with picosecond excitation studies, is caused by the presence of electrostriction. However, the variations of $n_2$
in the infrared region is to be interpreted as due to dispersion.

A discussion of the dispersion has been presented and several appropriate mechanisms are proposed. The experimental results show that the nonlinear refraction first increases with frequency and then decreases. If the observed dispersion is caused by the electrostriction, which is the dominating nonlinearity, it should increase without decreasing, for we are in the region of dispersion of the ultrasonic waves, where the speed of sound increases with frequency. If the dispersion is caused by the Kerr effect or field-induced absorption, the nonlinear refractive index is expected to increase toward the region of higher absorption. These results, therefore, indicate that more than a single microscopic process is responsible for the dispersion observed in this work. Experiments are suggested which permit the isolation of a single nonlinear process for future studies.
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APPENDIX A

POLARIZATION DEPENDENCE OF ABSORPTION AND LASER INDUCED SURFACE DAMAGE THRESHOLD IN WIRE GRID POLARIZERS

Microscopic wire grids deposited on a dielectric grating at a grazing angle are useful polarizers. They are compact, easily rotatable, can be used with converging and diverging beams, and work with all wavelengths at least four times smaller than the period of the grid, as long as the substrate's transmission permits. Gratings with a period as small as 0.4 μm are commercially available. With proper choice of the substrate, wire grid polarizers can be fabricated for use at any wavelength from 2 μm to far infrared. However, their principal limitation for use with high power lasers is their damage properties. In this work we present the results of measurements of the laser induced surface damage threshold and absorption of wire grid polarizers as a function of polarization and wavelength. A theoretical analysis of the absorption of wire grid polarizers versus orientation and wavelength will be presented based on a classical model for the wire grids under plane wave radiation which is in good agreement with the experimental results.
The Experiment

The polarizers used in our experiments were two wire grids of aluminum deposited on ZnSe substrate, one of the samples was overcoated with an antireflection (AR) coating on both surfaces. (Wire grid polarizers used in this work were acquired from PTR Optics Corporation, 145 Newton, Waltham, MA, 02454.) The period of wire grids was 0.84 μm with a symmetric 90° sawtooth groove shape, as illustrated schematically in figure A.1. The measurements were performed using linearly polarized, normal incident pulses from a hybrid CO₂ TEA laser. A complete description of the experimental set up is given in reference 1. The laser was operated in the TEM₀₀ spatial mode and single longitudinal mode with 130 ns (FWHM) pulses. Figure A.2 shows a pinhole beam scan of the spatial profile of the beam as well as a temporal profile detected by a photon drag detector (500 MHz response). The beam scan was performed just before a lens (F=25.4 cm) which focused the beam on the wire grid to a beam size of 0.36 mm (FWHM). Each site was irradiated only once (one-on-one measurement). Figure A.3 shows the experimental results for the sample with no AR coating for orientation of the wires parallel and perpendicular to the incident electric field. The damage threshold for this sample was 2 J/cm² (corresponding to a peak-on-axis irradiance of 15 MW/cm²) when the wires were parallel to the incident electric field.
Figure A.1—The surface structure of the wire grid and the model used for the calculations.
Figure A.2—The 200 μm pinhole beam scan performed just before the focusing lens ($F = 25.4$ cm). The inset shows the temporal profile of the pulse.
Figure A.3—Damage data for the wire grid without AR coating. Each point represents a single shot. Open circles are for polarization perpendicular to the wires. Closed circles are for polarization parallel to the wires.
Here we have defined the damage threshold as the lowest incident fluence (or irradiance) for which damage occurred. Occurrence of the damage was determined visually by observing a significant increase in the scattered light from a He-Ne laser propagating through the focal volume immediately after permanent changes were made in the sample by the CO$_2$ pulse. The relatively large overlap region for this sample was because of nonuniformities of the surface structure.

Figure A.4 shows the results for the sample with AR coating on both sides. The damage threshold for this sample was 0.8 J/cm$^2$ (6 MW/cm$^2$) when the wires were parallel to the incident polarization. The damage threshold of both samples increased by more than a factor of two when the wires were oriented perpendicular to the incident electric field (i.e., the orientation for maximum transmission). Table VI summarizes these results.

In order to study the effect of orientation of the wires on the absorption of the wire grids the photoacoustic method was used. We monitored the photoacoustic signal generated in the wire grid by coupling a transducer to the side of the sample. In order to avoid surface damage, the beam size on the front surface of the sample was kept large (3.2 mm FWHM). Figure A.5 shows the photoacoustic signal as a function of incident power for both orientations. The absorption of wire grids was greater by a factor of 3.6 when the polarization was parallel to the wires. This is
Figure A.4—Damage data for the wire grid with AR coating. The threshold for this sample was less than the sample without AR coating by more than a factor of two.
Figure A.5—Absorption of the wire grid at 10.6 μm. The photoacoustic signal indicates greater absorption when the wires are parallel to the polarization of the incident electric field.
TABLE VI
LASER INDUCED SURFACE DAMAGE THRESHOLD
OF THE WIRE GRID POLARIZERS

<table>
<thead>
<tr>
<th>Wire Grid</th>
<th>OPEN(∥) J/cm²</th>
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<td>AR Coated</td>
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<td>0.8</td>
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</tr>
<tr>
<td></td>
<td>15 MW/cm²</td>
<td>6 MW/cm²</td>
<td>2.5</td>
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<tr>
<td></td>
<td>36 MW/cm²</td>
<td>15 MW/cm²</td>
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</table>

consistent with the measurement of damage thresholds in parallel and perpendicular orientation. It is well known that the optical properties of wire grids are strongly dependent on the ratio of wavelength to the period of wires (λ/d). The wire grids are effective polarizers only when λ>>1. Both polarizations tend to be equally transmitted or reflected for λ/d close to or smaller than one. We used 30 ns pulses from a Q-switched YAG laser at 1.06 μm and its second harmonic (0.53 μm) to change λ/d in our experiment. Figure A.6 shows the photoacoustic signal versus incident power at 1.06 μm for both orientations. The absorption is smaller by a factor of 0.6 when the wires are parallel to the electric field. Figure A.7 is the result of the same
Figure A.6—Absorption of the wire grid at 1.06 μm. The photoacoustic signal indicates greater absorption when the wires are perpendicular to the absorption of the incident electric field.
Figure A.7—Absorption of the wire grid at 0.53 μm. The photoacoustic signal indicates greater absorption when the wires are perpendicular to the absorption of the incident electric field.
measurement at 0.53 μm. It shows that absorption is smaller by a factor of 0.8 when the wires are parallel to electric field. These results are summarized in Table VII.

**TABLE VII**

**RATIO OF ABSORPTIONS AS A FUNCTION OF WAVELENGTH**

| Pulsewidth FWHM ns | Spot Size FWH 2 M mm | Wavelength μm | Abs. (||)/Abs. (⊥) |
|---------------------|----------------------|---------------|--------------------|
| 130                 | 3.2                  | 10.6          | 3.6                |
| 30                  | 2.2                  | 1.06          | 0.62               |
| 20                  | 0.9                  | 0.53          | 0.82               |

**The Theory**

The theory of operation of wire grid polarizers has been the subject of investigations by many researchers in the past. All theoretical treatments start by assuming an ideal cross section for the wires. Flat strips and cylindrical wires have been considered. The effect of nonuniformities in the wire cannot be easily treated. Only the plane wave approximation has been considered. The problem of incident electric field with polarizations other than parallel to the wires, including ohmic loss has not been considered before. The reflected wave is regarded as originating from the currents induced in the wires, while the transmitted wave is the superposition of the induced and incident fields. Absorption is considered as the difference between incident
and sum of transmitted and reflected energies. The currents in the wires all have the same magnitudes (for plane wave illumination), but differ in phase from wire to wire. Only Casey and Lewis\textsuperscript{6,7} have considered the effects of ohmic loss (i.e., finite conductivity of the wires) on the transmission and reflection of these devices, however, their calculations were limited to the case of the light polarization vector parallel to the wires. Here we consider the cases of polarization vector parallel and perpendicular to the wires and calculate the ratio of the absorption for the two cases as a function of $d/\lambda$.

Following the treatment in reference 5 we model the wires as flat strips of conductors on a flat insulating substrate, as shown in figure A.1. The parallel strips are assumed to have conductivity $\sigma$ and thickness $h>>>\delta$ where $\delta$ is the wavelength dependent skin depth. The width of the wires is taken to be equal to their separation $d$. We assume linearly polarized plane wave at normal incidence. The dissipated power density in the conducting strips is $W=|J_{\text{eff}}|^2|\text{Re}(Z)|$; where $J_{\text{eff}}=\int_0^\infty J(x)dx$ is the effective surface current density induced in the wires,\textsuperscript{8} $Z$ is the impedance of the wires and $J(x)$ is the current at a distance $x$ inside the metal strip. We first calculate the impedance of a conducting strip for polarization parallel to the strips to be $Z = (1+i)/\sigma\delta$. This is the same as the surface impedance of a good conductor. In the case of polarization perpendicular
to the strips, this impedance is added to the impedance of the effective capacitance per unit length $C$ formed by the space between the conducting wires (see figure A.1). If $\omega$ and $c$ are the frequency and the speed of incident light, we have

$$Z_J = Z_{||} + \frac{1}{i\omega c},$$
or

$$Z = \frac{1}{\omega^2} + i\left(\frac{1}{\omega^2} - \frac{\lambda}{2\pi cC}\right).$$

The quantity $C$ is dependent on the geometry of the problem.

The total conduction current density in both orientations are

$$J_{eff}^\parallel = \frac{E(0)}{Z_{||}};$$
and

$$J_{eff}^\perp = \frac{E(0)}{Z_{\perp}}.$$ Here, $E(0)$ is the total field on the surface. The ratio of absorption of the radiation in the conducting strips in the parallel and perpendicular orientation is

$$R = \frac{W_{||}}{W_{\perp}} = \frac{Z_{\perp}}{Z_{||}} = \frac{1}{2} \left[ 1 + \frac{\lambda}{\pi c C} \sqrt{\left(\frac{1}{\pi} \frac{\lambda^3}{\pi^3 c^3}\right)^2} \right].$$

As a first approximation we calculate $C$ to be

$$C = \eta \sqrt{\frac{\lambda}{\pi \mu c}};$$
where $\eta$ is a unitless geometric factor and is treated as a free adjustable parameter. The ratio of the absorptions now becomes

$$R = \frac{1}{2} \left[ 1 + \frac{\lambda^3}{4\pi c \eta} \right]^2.$$ Figure A.8 shows the plot of $R$ as a function of wavelength and the results of measurements of this ratio for three
Figure A.8—Plot of the ratio of the absorption of parallel polarized light to the absorption of perpendicular polarized light versus wavelength. The solid curve is the theoretical calculation of the ratio, and the points are data from this work.
wavelengths. The best fit is obtained with \( \eta = 124.8 \). In our model the electromagnetic parameters \( \epsilon, \mu, \sigma \) are considered as their static values. The resonance frequencies of the bound electrons of metallic atoms are known to lie far in the violet or ultraviolet region of the spectrum. Therefore the inductive capacity of a metal, \( \epsilon \), in the visible red can safely be assumed equal to its static values. Moreover if the conductor is nonferromagnetic the permeability is also approximately equal to its static value. It was also demonstrated by Hagen and Rubens\(^9\) that the values of conductivity measured under static conditions may be employed without appreciable error well into the infrared. Only at wavelengths shorter than about 0.25 \( \mu \text{m} \) the conductivity exhibits a dependence on frequency. In our experiments wavelengths are longer than 0.53 \( \mu \text{m} \). We used the value of static conductivity of \( 1.55 \times 10^{17} \text{ s}^{-1} \) for aluminum given in reference 10. The agreement between theory and experiment is remarkable considering the crude model used to calculate the capacitance of the grid.

**Summary**

We have measured the surface damage thresholds of wire grid polarizers at 10.6 \( \mu \text{m} \). Damage thresholds were maximum when the polarization of light was perpendicular to wires. The ratio of absorption of parallel polarized light to the absorption of perpendicular polarized light depends on \( \lambda/d \).
The ratio of these two absorptions is greater than one for \( \lambda/d \gg 1 \) and it is less than one for \( \lambda/d \) close to or smaller than one. This behavior is predicted by the theoretical model presented in this work. The ratio of surface damage threshold under perpendicular polarized light to that under parallel polarized light at 10.6 \( \mu \text{m} \) is consistent with the ratio of absorptions for the two orientations.
CHAPTER BIBLIOGRAPHY


APPENDIX B

THE NUMERICAL TECHNIQUE FOR SOLUTION OF
THE NONLINEAR WAVE EQUATION

In Chapter III the beam distortion measurement technique was successfully used to measure the nonlinear refractive index of CS$_2$. This is an accurate and sensitive method provided that the nonlinear wave equation can be solved. This appendix is intended for the interested reader who wishes to reproduce our simulations of the nonlinear propagation, or has a similar problem in mind. Since a meaningful result can not be obtained from a computer program without understanding the way it operate, the method used will be considered in some detail. After stating the problem the considerations which are taken into account in selection of a numerical method will be discussed. The method used to obtain the results of Chapter III, with its advantages and disadvantages, will be described. The anatomy of the actual code used in this work will be discussed and finally a list of the program will be presented.

The Nonlinear Wave Equation

We wish to solve the nonlinear wave equation

\[ \frac{\partial E}{\partial z} = -i/2k(\nabla^2 E + A|E|^2) - \alpha E/2 \]
for the electric field. Here, $E(r,z)$ is the complex electric field, $r$ the radial coordinate, $z$ the propagation distance, $k$ the wave number in the medium, and $\alpha$ the linear absorption coefficient. The nonlinearity lies in the cubic term and its complex coefficient is given by

$$A = k^2 n_2 / n_0 - i \kappa n_0 \epsilon_0 \beta / 2.$$ 

Where, $n_2$ is the nonlinear refractive index and $\beta$ is the two photon absorption coefficient. In arriving at this scalar second-order partial differential equation certain approximating steps have been taken. The medium is assumed to be isotropic. The electric field is perpendicular to the propagation direction ($z$). (For a circularly polarized light similar equation can be written for both components of the electric field.) The term $\nabla(\nabla \cdot E)$ is assumed to be negligible. Only first and third order susceptibilities are included. The induced polarization in the material is assumed to respond instantaneously to the variations in the incident field. The field amplitude is assumed not to vary appreciably over a distance of a wavelength (i.e., slowly varying envelop approximation). Assumption of the circular symmetry for the beam profile throughout the propagation in the sample is also embedded in the form of the equation. We have already justified some of these assumptions. However, all of these approximations are invoked commonly in the literature for similar experimental conditions.$^{1,2}$ Therefore
we do not duel on a rigorous justification of these assumptions in this discussion. This parabolic differential equation is nothing but the nonlinear Schrödinger equation and there is no known analytic solution to the three dimensional form of it given above. The two dimensional equation, however, has been solved exactly by Zakharov. Unfortunately his soliton solutions can not be applied to a real three dimensional problem at hand. It has been shown that analytic solutions can be obtained by adding the assumption that the nonlinear sample is thin enough such that the intensity variations of the beam in the z direction is negligible. Assuming such a thin sample allows one to drop the transverse laplacian from the equation. This assumption together with constant shape approximation is used in reference 5. The behavior of the solutions of the wave equation under many different approximations is reviewed in references 6 and 7. The equation under constant shape approximation was solved numerically by Wagner. Wang by using numerical methods solved the equation without ignoring the $\nabla(\nabla \cdot E)$ term. His results were in agreement with Kelly who had earlier solved the equation numerically ignoring this term. Marburger and Dyshko have also independently solved the same equation numerically. The numerical methods used by these authors were to transform the differential equation to an explicit difference equation then solve that
step by step (i.e., explicit finite difference method\textsuperscript{13}) or to reduce the differential equation to a system of simultaneous equations (i.e., implicit finite difference method\textsuperscript{13}). Kelly and Marburger used the method which was developed by Harmuth\textsuperscript{14}. This is a second order explicit difference scheme with the truncation error of the order of $h^3$; where $h$ is the step size taken in the propagation direction. The value of $dE/dz$ at any point, $z$, is calculated by using the values of $E$ at the two points of $z-h$ and $z+h$. The theoretical analysis of Harmuth shows that the stability of the method is not very good. Marburger in his implementation of the method artificially enforced the stability of the method by replacing one of the electric fields in the cubic term at the position $z$ with the average of the values of the electric field at the positions of $z-h$ and $z+h$. The same approximation was also made for the linear absorption term. Dyshko did not make these adjustments instead he used a second order implicit difference scheme which was shown to be stable. In this method the solution is contained implicitly in a system of simultaneous equations which can be solved with standard methods. Recently a finite Hankel transform technique\textsuperscript{15,16} and the so called split-step Fourier technique\textsuperscript{17} have been claimed to be advantageous for numerical solutions of the nonlinear wave equation with cubic term.
The Numerical Method of Solution

Different methods were used to solve the nonlinear wave equation on a NAS 8083 computer. By assuming the complex electric field of the form \( E(r,z) = A(r,z) \exp[iF(r,z)] \) the equation was reduced to two coupled differential equations for real amplitude \( A \) and real phase \( F \). These equations were then solved simultaneously by using the method of lines. In this method the function \( E(r,z) \) is defined at mesh points in the \( r \) coordinate but it is kept as a continuous function in the \( z \) coordinate (i.e., \( E_i(z) \) for \( i = 1, 2, \ldots, N \)). The original differential equation is reduced to a system of \( N \) ordinary differential equations for \( E_i \) which is solved by an implicit multistep method. The coupled equations for the real amplitude and the phase were solved using the available code in the IMSL library. The program DPDES in the IMSL library is designed to solve a system of partial differential equations by the method of lines. The method was successful in solving the equation with great accuracy when the sample length was much smaller than the Rayleigh range. However, the program encountered singular matrices in solving the resulting system of algebraic equations when the sample thickness was increased. Some improvements were obtained when the division of the original equation into two coupled equation for real amplitude and phase was abandoned in favor of defining the electric field in terms of its real and
imaginary parts [i.e., \( E(r,z) = E_r(r,z) + i E_i(r,z) \)]. Nevertheless the generalized differential equation solver proved to be inefficient and limiting for as a closed package it offered little information about the inner-processes which are required for tuning the program to a specific problem for maximum efficiency and accuracy. Next an iterative predictor-corrector method was used. A program was developed based on the Adams-Moulton\(^{20}\) predictor formula and Diels\(^{21}\) corrector formula. These are both of fourth order accuracy with the truncation error of the order of \( h^5 \). The transverse laplacian was calculated using a five point formula.\(^{22}\) This method can be summarized in a four step recipe:

1. Use \( \frac{dE}{dz} \) at four previous steps with predictor formula to predict the value of \( E \) at \( z \).
2. Use the predicted \( E \) with the differential equation to find \( \frac{dE}{dz} \) at \( z \).
3. Use the predicted \( E \) and the values of \( \frac{dE}{dz} \) at the five positions with the corrector formula to find corrected \( E \) at \( z \).
4. Replace the predicted value with the corrected value of \( E \) and iterate steps 2 to 4 until there is no difference between the predicted and corrected values.

The values of the derivatives at the first four steps must be provided by some other means for the program. In this
implementation these four values were simply put equal to zero. The method solved the equation for thick samples. The results were in good agreement with Marburger's results and the results obtained by the method of lines for thin samples. The solution to the linear propagation could be obtained with extreme accuracy (less than 0.01% difference with the analytical results). The agreement with the approximate results in the case of thin samples was excellent. The method is stable and accurate but because of its iterative nature it requires a long computation time. Most of the computation time is consumed in iterative process of correcting for the large initial error in the predicted value. Hamming*23-25 studied the stability of a large family of predictor corrector formulas. He showed that the truncation error in the predictor and corrector formulas can be estimated and compensated for by using the difference of the predicted and corrected values in the previous step. The iteration can therefore be abandoned leading to greater computational speed without loss of accuracy. The recipe for this method is given in five steps.

1. The value of $E$ at $z$ is predicted using $dE/dz$ at $z-h$, $z-2h$, $z-3h$, and the value of $E$ at $z-4h$.
2. The predicted value is modified using the difference between the predicted and corrected $E$ in the previous step which is a measure of truncation error committed in step 1 above.
3. The modified value is used in the differential equation and $dE/dz$ at $z$ is calculated. (Note, that the bulk of calculation time is spent in this step.)

4. The value of $E$ at $z$ is corrected using the modified $dE/dz$ and the $dE/dz$ at $z-h$ and $z-2h$ in addition to the values of $E$ at $z-h$ and $z-3h$.

5. The final value of $E$ at $z$ is calculated by modifying the corrected value of $E$ from step 4 above.

The actual formulas for the predictor, modifier, corrector, and the final result can be found in the listing of the program at the end of the next section. In this implementation the starting values were obtained by a fourth order Ronge-Kutta method\textsuperscript{25} which has the same accuracy of the predictor corrector method but it needs four times more calculation for each step, instead it is a self-starting technique. The Hamming's method described above has a large margin of stability and has the advantage that the information about the truncation error is embedded in the difference between the predicted and corrected values at any step. This information can be used to adjust the step size $h$ for maximum efficiency while the program is running. This method was used successfully to obtain the results in Chapter III.
The Fortran program which is listed at the end of this section solves the nonlinear wave equation describing self-focusing. The solution of the wave equation is then propagated beyond the nonlinear sample to the position of the detector by evaluating the Fresnel integral. Its inputs are the experimental conditions such as the wavelength, incident power, index of refraction, absorption, etc., in addition to the geometrical parameters such as the sample length, position of the focus, position of the detector, beam size, etc. Its outputs are intensity and phase profile at any point in the sample and at the detector position, intensity as a function of z at any radial point, real and imaginary parts of the electric filed, and some other diagnostic parameters. All of the outputs are optional. In the case of our experiments the only output needed was the beam profile at the position of the detector. All other informations provided by the program are treated as diagnostics to test the performance of the program under specific conditions and they can be by-passed to save time once the program is tuned for maximum efficiency. There are 13 subroutines in addition to the main part of the program. A brief description of each subroutine will follow.

Subroutine FIRST calculates the electric field at $z = 0$ using the information supplied by the user. Intensity and
phase profiles are also calculated. In this implementation an initial gaussian profile is assumed but any profile can be supplied. If the profile is available in the form of data points, rather than function, it should be provided to this subroutine. This subroutine is called only once at the beginning of the program.

Subroutine OUTZ provides information as requested while the program is taking steps in z direction. Presently these informations are on axis intensity, real and imaginary electric field, power of the beam, estimated truncation error, and the step size.

Subroutine OUTR provides radial profiles as requested. This subroutine is called once at the end of the sample and once at the position of the detector but it can be called at any position in the sample and as many times as necessary. It provides intensity and phase profiles as well as the real and imaginary parts of the electric field.

Subroutine PHASE provides the on axis phase of the beam for the subroutine OUTZ. Examination of phase profile at the end of the sample given by OUTR is an easy way of making sure that the program is operating properly. The phase is defined by the angle of the electric field vector in the complex plane with the real axis. However, as the program is stepping in z the electric field is rotating in the complex plane and every time it crosses the imaginary axis there
appears a jump in the phase profile. This subroutine detects these discontinuities and corrects them by appropriately adding or subtracting a $\pi$ to or from the phase.

Subroutine PROPZ is the nucleus of the program. The formulas for the predictor, modifier, and corrector are given here. The decision on whether to double the step size or half the step size is taken in this subroutine.

Subroutine START is responsible to provide the values of $E$ and $dE/dz$ for the first four steps.

Subroutine RUNG is the nucleus of START. This subroutine takes one step in $z$ with the Rung-Kutta method. It has the same accuracy (fourth order) as the Hamming's method. In principal the whole problem can be solved with this subroutine if there is enough computer time.

Subroutine DIFEQ contains the differential equation. The transverse laplacian and $dE/dz$ is calculated in this subroutine. In the Hamming method this subroutine is called only once at every step, whereas in the Rung-Kutta method DIFEQ is called four times at every step.

Subroutine INTERP calculates $E$ and $dE/dz$ at midpoints. This is done when it is decided that there is too much truncation error and the step size must be cut in half. When there is too much accuracy the step size is doubled in order not to waist time. In this case subroutine START is called to provide a new set of starting values at that position.
ALP1 and ALP2 are the upper and lower limit on the truncation error and should be supplied by the user. If ALP1 is set equal to ALP2 the program will continue with the same step size. This will save time in cases when a single step size is good for the whole propagation range. The problem of round off errors is solved by doing the calculations in double precision.

Subroutine POWER calculates the power in the beam. It is correctly suggested by Marburger\textsuperscript{6} that even with zero linear absorption the beam may loose power as it propagates. This is mainly because of diffraction and the fact that the beam profile is defined in a finite mesh. The beam may diffract out of the definition mesh or be focused so hard that only a few mesh points effectively define the beam profile. These effects can be best detected by monitoring the power of the beam. This subroutine provides the power when requested. This information could be used to adjust the mesh size automatically to fit the size of the beam.

Subroutine FRENEL calculates the Fresnel integral and gives the intensity profile at the position of the detector. The integration scheme is also of fourth order accuracy.

Subroutine CALCON calculates the constants used in the five point formulas. These are done separately to save time.

Subroutine CALDZ calculates the coefficients of the predictor corrector formulas. When the step size changes these must be calculated again.
The fact that the computer does not give any error messages is not enough reason to believe the output of the program. Before accepting the results from the program many tests must be performed. For all the results presented in Chapter III these tests were performed. First the number of radial mesh points and the step size is determined that gives reasonable results. Then upper and lower bound on the truncation error is determined. This is done by studying the behavior of the truncation error as the step size is changed. The parameter ABSPC in the program is the absolute value of the sum of the estimated truncation errors in calculating all the profile at z. A plot of the electric field in the complex plane as the program steps in z is very helpful in determining whether or not the truncation error is acceptable. By putting the linear absorption equal to zero the variation of power is studied. Changes of less than 0.1% are tolerated. The control parameter for this is the number of radial mesh points. Variation of this parameter has the side effect of changing the optimum step size. The results of the program with zero nonlinearity is compared with the analytic solutions. In this regard the phase profile is much more sensitive to instabilities and errors than the intensity profile. The test for the linear absorption term is also made by comparing the on axis intensity with the Beer's law. The results of the program with nonlinearity under thin
sample condition is compared with the approximate solutions. It is with such care that the output of the program can be compared with the experimental results.

In view of the importance of a general purpose program which can solve the nonlinear propagation problem some improvements to this code can be suggested. In general the results of the program are not reliable beyond singularity. The beam collapses to such a small space that can not be well defined with a coarse mesh. The natural solution to this problem is to have an adjustable mesh which changes according to the beam size. Preliminary numerical experiments have shown promising results. In order to model the pulse shape more accurately in a reasonable time there is no other solution but to use a parallel processor or a supercomputer. Still other numerical methods can be explored.
THE OUTPUT OF THIS PROGRAM CAN BE TRANSFERRED TO IBM-AT.

THIS PROGRAM IS CALLED "MAXHAM". IT SOLVES WAVE EQUATION FOR COMPLEX ELECTRIC FIELD. THE EQUATION CONTAINS FOUR TERMS:

1-1ST DERIVATIVE W.R.T Z
2-TRANSVERSE LAPLACIAN
3-LINEAR ABSorption
4-CUBIC NONLINEARITY (i.e., 2 PH.ABS., AND S.F.)

THE NUMERICAL METHOD IS 4TH ORDER MODIFIED HAMMING PREDICTOR CORRECTOR WITH THE TRUNCATION ERROR OF ORDER DZ**5. THE TRANSVERSE LAPLACIAN IS COMPUTED BY A SET OF FIVE POINT FORMULAS.

ALL CALCULATION ARE IN DOUBLE PRECISION

C

COMPLEX*16 EO(200),E1(200),E2(200),E3(200),D1(200),
  D2(200),D3(200)
COMPLEX*16 PC(200),E31(200),RK
REAL*8 R(200),HWEM,WAVLEN,SAMPL,ZDET,ABS0,ABS2,DX0,DX2
1,DZ,DR
REAL*8 PI,SPDL,COF,WAVNUM,WNK,RI,AIL,FF1,EPE,EIM,ERE1,
1DET
REAL*8 AMP,PHS,RI(200),S,ZCH,Z,ABSPC,ALP1,ALP2
REAL*8 PWR,WO,DL,EPSON,ZO,RO,HI
INTEGER*4 I,J,NR,IOUT,IICHK,NZOUT,NROUT,NFRENL
COMMON/BLOCK/ ERE1,DET,PHS,COF,IOUT,IICHK
COMMON/FACTOR/ ABSPC,ALP1,ALP2
COMMON/RADIOS/ R,DR,NR
COMMON/FIELDS/ EO,E1,E2,E3,D1,D2,D3,PC
COMMON/SPACE/E31,R1
COMMON/ONLY/ ABSO

OUTPUT CONTROL INFORMATION

NZOUT=100
IOUT=1
NROUT=0
NFRENL=0

-NZOUT IS # OF OUTPUTS IN Z ; 0,1,2,...NZ
-IOUT IS RADIAL INDEX FOR NZOUT ; 1,2,...NR
-NROUT IS 1 IF WANT PROFILE AT SAMPL, 0 IF NOT.

-NFRENL IS 1 IF WANT PROFILE AT ZDET, 0 IF NOT.

ALL UNITS ARE IN CGS(ESU), BUT POWER, WHICH IS IN KW

PWR = 65.
W0 = 1.413D-2
DL = 0.0
WAVLEN = 10.5D-4
SAMPL = 2.2
ZDET = 0.20D2
ABS0 = 0.17
ABS2 = 0.0
DX0 = 0.16D1
DX2 = 9.2D-10
NZ = 10000
NR = 121
PI = 3.14159265359
SPDL = 2.99792458D10
EPS0 = 7.9574D-2
DUMMY = 1.0

Z0 = PI*W0**2*DX0/WAVLEN
HWEM = DSQRT(W0*W0*(1.+DL*DL/Z0**2.)/2.)

IF (DL.EQ.0.) GO TO 5
R0 = DL*(1.+Z0*Z0/DL**2)
GO TO 6

5
R0 = 1.
DUMMY = 0.

6
ALP1 = 0.1E-1
ALP2 = 0.1E-3

IICHK = 11

WRITE (6, 10) PWR, W0, WAVLEN, SAMPL, ZDET
WRITE (6, 11) DX0, DX2, ABS2, ALP1, ALP2
WRITE (6, 12) NR, NZ, NZOUT, NROUT, NFRENL

COF = DSQRT(8.0*PI/(DX0*SPDL))
WNK = 2.0*PI/WAVLEN
WAVNUM = 2.0*PI*DX0/WAVLEN
RK = WAVNUM**2*DX2/DX0-(0., 1.)*WAVNUM*SPDL*EPS0*DX0*ABS2/2.

HI = PWR*1.D10/(PI*HWEM**2)
DZ = SAMPL/NZ
DR = HWEM*4./ (NR-1)
CALL CALCON (DR)

-> START OF THE LOOP <-
CALL CALDZE(DZ)
CALL FIRST(E0,HI,HWEM,WAVNUM,DUMY,R0,DL,Z0)
   Z = 0.0
   ABSPC = 0.0
IF(NZOUT.EQ.0) GO TO 20
CALL POWER(E0,P,DZ)
CALL OUTZ(E0(IOUT),Z,P,DZ)
20  CALL START(Z,DZ,RK,WAVNUM)
      J = 1
111  CALL PROPZ(Z,DZ,RK,WAVNUM)
CALL PHASE(E3(IOUT),ERE,EIM)
IF(DZ.LT.1.D-10) GO TO 112
   ZCH = Z+DZ
IF(ZCH.GE.SAMPL) GO TO 112
IF(NZOUT.EQ.0) GO TO 111
   S=J*SAMPL/NZOUT
IF(ZCH.LT.S) GO TO 111
      J = J+1
CALL POWER(E3,P,DZ)
CALL OUTZ(E3(IOUT),Z,P,DZ)
GO TO 111
112  IF(NZOUT.EQ.0) GO TO 40
   CALL POWER(E3,P,DZ)
   CALL OUTZ(E3(IOUT),Z,P,DZ)
40  IF(NROUT.EQ.0) GO TO 50
   CALL OUTR(E3,R,NR)
50  IF(NFRENL.EQ.0) GO TO 60
   CALL FRENEL(E3,WNK,WAVLEN,ZDET,W0)
   DET = 0.0
   CALL OUTR(E31,R1,NR)
60  CONTINUE
STOP
END

C------------- INITIAL PROFILES OF E(REAL),E(IMAG.) USING C
C INTENSITY AND PHASE PROFILE. C
C
SUBROUTINE FIRST(E0,HI,HWEM,WAVNUM,DUMY,R0,DL,Z0)
COMPLEX*16 E0(200)
REAL*8 R(200),DR,All,HI,HWEM,FF1,WAVNUM,R0,DL,Z0
REAL*8 ERE,EIM,COF,ERE1,DET,PHS
INTEGER*4 NR,I,IOUT,IICHK
COMMON/BLOCK/ ERE1,DET,PHS,COF,IOUT,IICHK
COMMON/RADIOS/ R,DR,NR
   DO 16  I=1,NR
      RI=I-1
      R(I)=DR*RI
   AII=HI*DEXP(-(R(I)/HWEM)**2)
   FF1=DUMY*WAVNUM*(DL-DATAN(DL/Z0)+R(I)**2/(2.*R0))
   27  ERE=COF*DSQRT(AII)*DCOS(FF1)
EIM = COF * DSQRT(A11) * DSIN(FF1)
E0(I) = ERE + (0.0, 1.) * EIM
CONTINUE
ERE1 = .5 * (DCONJG(E0(IOUT)) + E0(IOUT))
DET = 0.0
RETURN
END

C --------------------------------------------------
C ALL OUTPUTS ARE IN (CGS).
C --------------------------------------------------

C SUBROUTINE OUTZ(E, Z, P, DZ)
C COMPLEX*16 E
REAL*8 R, Z, PHS, AMP, ERE, EIM, ERE1, DET, COF, INTENS
REAL*8 P, DZ, ABSPC, ALP1, ALP2
INTEGER*4 IOUT, IICHK
COMMON/BLOCK/ ERE1, DET, PHS, COF, IOUT, IICHK
COMMON/FACTOR/ ABSPC, ALP1, ALP2
ERE = .5 * (DCONJG(E) + E)
EIM = (0.0, 1.0) * (ERE - E)
AMP = DSQRT(ERE**2 + EIM**2)
INTENS = AMP**2 / COF**2
WRITE(6,5) Z, INTENS, ERE, EIM, P, ABSPC, DZ
5 FORMAT(F9.5, 3(1X, E12.5), 1X, F9.4, 1X, E12.5, 1X, F8.6)
RETURN
END

C --------------------------------------------------
C OUTPUT THE PROFILE.
C --------------------------------------------------

C SUBROUTINE OUTR(E, R, NR)
C COMPLEX*16 E(200)
REAL*8 R(200), ERE, EIM, PHS, AMP, ERE1, DET, COF, INTENS
INTEGER*4 NR, IOUT, IICHK
NRR = NR / 2
62 DO 70 J = 1, NR
CALL PHASE(E(J), ERE, EIM)
AMP = DSQRT(ERE**2 + EIM**2)
INTENS = AMP**2 / COF**2
WRITE(6,65) R(J), INTENS, PHS, ERE, EIM
65 FORMAT(5E15.7)
70 CONTINUE
RETURN
END

C --------------------------------------------------
C THIS SUBROUTINE MONITORS THE EVOLUTION OF PHASE ON THE AXIS, IN ORDER TO AVOID DISCONTINUITIES OF THE ORDER OF INTEGER*PI.
C --------------------------------------------------

C SUBROUTINE PHASE(E, ERE, EIM)
C COMPLEX*16 E
REAL*8 ERE, EIM, ERE1, DET, PHS, PI, COF
INTEGER*4 IOUT, ISE1, ISERE, ISEIM, NA, NB, IICHK
COMMON/BLOCK/ ERE1, DET, PHS, COF, IOUT, IICHK
PI = 3.14159265359
ERE = .5*(DCONJG(E)+E)
EIM = (0.0, 1.0)* (ERE-E)
ISE1 = 1
IF (ERE1.LT.0.) ISE1 = -1
ISERE = 1
ISEIM = 1
IF (EIM.LT.0.) ISEIM = -1
IF (EIM.EQ.0.) ISEIM = 0
NA = ISE1*ISERE
NB = ISE1*ISEIM
IF (NA.GT.0) GO TO 5
DET = DET + PI*NB
5
    PHS = DATAN(EIM/ERE) + DET
    ERE1 = ERE
    RETURN
END

C------------------------ THIS IS THE NUCLEUS OF THE PROGRAM C
C
SUBROUTINE PROPZ(Z, DZ, RK, WAVNUM)
COMPLEX*16 E0(200), E1(200), E2(200), E3(200), D1(200),
                 D2(200), D3(200)
COMPLEX*16 PC(200), RK, EMOD(200), DMOD(200), PRE(200),
1CORE(200)
REAL*8 R(200), Z, DZ, DR, WAVNUM, ALP1, ALP2, ABSPC, ERE1,
1DET, PHS, COF
REAL*8 ALP1, ALP2, A1, A2, A112, B9, B1, B3, B6, B121
REAL*8 A112, B9, B1, B121, A242, A16, A13, DR12, DRS12, DRS6
REAL*8 A1, A2, B3, B6, A3, A9, DZ15, DZ90, DZ3, DZ54, DZ27
INTEGER*4 I, NR, IOUT, IICHK
COMMON/BLOCK/ ERE1, DET, PHS, COF, IOUT, IICHK
COMMON/FACTOR/ ABSPC, ALP1, ALP2
COMMON/FIELDS/ E0, E1, E2, E3, D1, D2, D3, PC
COMMON/RADIOS/ R, DR, NR
COMMON/CNSTNT/ A112, B9, B1, B121, A242, A16, A13, DR12,
1DRS12, DRS6
COMMON/DZFACT/ A1, A2, B3, B6, A3, A9, DZ15, DZ90, DZ3,
1DZ54, DZ27
5
    ABSPC = 0.0
    DO 10 I = 1, NR
         PRE(I) = E0(I) + A2*(D3(I) + D1(I)) - A1*D2(I)
         EMOD(I) = PRE(I) - A112*PC(I)
    CONTINUE
    CALL DIFEQU(EMOD, DMOD, DZ, RK, WAVNUM)
    DO 20 I = 1, NR
CORE(I) = B9*E3(I) - B1*E1(I) + B3*(DMOD(I) - D2(I)) + B6*D3(I)  
PC(I) = PRE(I) - CORE(I)  
CORE(I) = CORE(I) + B121*PC(I)  
ABSPC = CDABS(PC(I)) + ABSPC

20 CONTINUE
IF(ALP1.EQ.ALPM2) GO TO 555
IICHK = IICHK + 1
IF((IICHK.GT.0).AND.(IICHK.LE.10)) GO TO 555
IF(ALP2.GT.ALP1) GO TO 100
IF(ALP2.LT.ALP1) GO TO 200
GO TO 555

100 CALL INTERP(DZ, RK, WAVNUM)
   DZ = DZ/2.
   CALL CALDZ(DZ)
   IICHK = 0
   GO TO 5

200 DZ = 2.*DZ
   CALL CALDZ(DZ)
   CALL START(Z, DZ, RK, WAVNUM)
   IICHK = 4
   GO TO 5

555 CALL DIFEQU(CORE, DMOD, DZ, RK, WAVNUM)
   DO 40 I = 1, NR
      E0(I) = E1(I)
      E1(I) = E2(I)
      E2(I) = E3(I)
      E3(I) = CORE(I)
      D1(I) = D2(I)
      D2(I) = D3(I)
      D3(I) = DMOD(I)

40 CONTINUE
   Z = Z + DZ
   RETURN
END

C   -------------------------------C
C   THIS SUBROUTINE CALCULATES THE STARTING VALUES.  C
C   -------------------------------C
SUBROUTINE START(Z, DZ, RK, WAVNUM)
   COMPLEX*16 EO(200), E1(200), E2(200), E3(200), D1(200),
   1 D2(200)
   COMPLEX*16 D3(200), PC(200), DO(200), RK
   REAL*8 R(200), Z, DZ, WAVNUM, DR, ERE1, DET, PHS, COF, ERE, EIM
   REAL*8 ABSPC, ZCHEK, ALP1, ALP2, A1, A3, A9
   REAL*8 A112, B9, B1, B121, A242, A16, A13, DR12, DR51, DR56
   REAL*8 A1, A2, B3, B6, A3, A9, DZ15, DZ90, DZ23, DZ54, DZ27
   INTEGER*4 I, NR, IOUT, IICHK
   COMMON/BLOCK/ ERE1, DET, PHS, COF, IOUT, IICHK
   COMMON/FACTOR/ ABSPC, ALP1, ALP2
   COMMON/FIELDS/ EO, E1, E2, E3, D1, D2, D3, PC
   COMMON/RADIOS/ R, DR, NR
COMMON/CNSTNNT/ A112, B9, B1, B121, A242, A16, A13, DR12, 
1DRS12, DRS6  
COMMON/DZFACT/ A1, A2, B3, B6, A3, A9, DZ15, DZ90, DZ3, DZ54, 
1DZ27  
25  ABSPC=0.0 
CALL DIFEQU(E0, D0, DZ, RK, WAVNUM) 
CALL PHASE(E0(IOUT), ERE, EIM)  
CALL RUNG(E0, E1, D1, Z, DZ, RK, WAVNUM) 
CALL PHASE(E1(IOUT), ERE, EIM)  
CALL RUNG(E1, E2, D2, Z, DZ, RK, WAVNUM)  
CALL PHASE(E2(IOUT), ERE, EIM)  
CALL RUNG(E2, E3, D3, Z, DZ, RK, WAVNUM)  
CALL PHASE(E3(IOUT), ERE, EIM)  
ZCHEK=4.*DZ  
DO 10 I=1, NR  
PC(I)=A242*(E3(I)-E0(I))-A3*(D3(I)+D0(I))-A9*(D2(I)+ 
1D1(I))  
ABSPC=ABSPC+CDABS(PC(I))  
10 CONTINUE  
IF(Z.GE.ZCHEK) GO TO 50  
IF(ALP1.EQ.ALP2) GO TO 50  
IF(ABSPC.LT.ALP2) GO TO 200  
IF(ABSPC.GT.ALP1) GO TO 100  
GO TO 50  
100 Z=0.0  
DZ=DZ/2.  
CALL CALDZE(DZ)  
GO TO 25  
200 Z=0.0  
DZ=DZ*2.  
CALL CALDZE(DZ)  
GO TO 25  
50 RETURN  
END  
C-----------------------------------------------C  
C USE RUNG-KUTTA METHOD TO START  
C-----------------------------------------------C  
SUBROUTINE RUNG(E0, E, D, Z, DZ, RK, WAVNUM)  
COMPLEX*16 E0(200), E(200), D(200), FK(4,200), RK  
REAL*8 R(200), Z, DZ, WAVNUM, DR, C(4), A1, A2  
REAL*8 A112, B9, B1, B121, A242, A16, A13, DR12, DRS12, DRS6  
INTEGER*4 I, J, NR  
COMMON/RADIOS/ R, DR, NR  
COMMON/CNSTNNT/ A112, B9, B1, B121, A242, A16, A13, DR12, 
1DRS12, DRS6  
C(1)=2.  
C(2)=2.  
C(3)=1.  
C(4)=1.  
DO 10 I=1, NR
E(I)=E0(I)

CONTINUE

DO 30 J=1,4
    CALL DIFEQU(E,D,DZ,RK,WAVNUM)
    DO 20 I=1,NR
        FK(J,I)=DZ*D(I)
        IF(J.EQ.4) GO TO 20
        E(I)=E0(I)+FK(J,I)/C(J)
    20 CONTINUE

30 CONTINUE

DO 40 I=1,NR
    E(I)=E0(I)+A16*(FK(1,1)+FK(4,1))+A13*(FK(2,1)+FK(3,1))
40 CONTINUE

CALL DIFEQU(E,D,DZ,RK,WAVNUM)
Z=Z+DZ
RETURN
END

C------------------------------------------------------------------------C
C DEFINE THE DIFFERENTIAL EQUATION HERE                                C
C------------------------------------------------------------------------C

SUBROUTINE DIFEQU(E,D,DZ,RK,WAVNUM)
COMPLEX*16 E(200),D(200),DDE(200),RK,D1E,D2E
REAL*8 R(200),DR,DZ,WAVNUM,ABSO
REAL*8 A112,B9,B1,B121,A242,A16,A13,DR12,DRS12,DRS6
INTEGER*4 I,II,12,13,14,15,IC,ICC,NR,NR1
COMMON/RADIOS/ R,DR,NR
COMMON/CNSTNT/ A112,B9,B1,B121,A242,A16,A13,DR12,DRS12,DRS6
COMMON/ONLY/ ABSO

DDE(1)=(16.*E(2)-15.*E(1)-E(3))/DRS6
D1E=(E(2)-8.*(E(1)-E(3))-E(4))/DR12
D2E=(16.*(E(1)+E(3))-31.*E(2)-E(4))/DRS12
DDE(2)=D2E+D1E/R(2)

NR1=NR+1
DO 2 I=6,NR1
    I1=I-1
    I2=I-2
    I3=I-3
    I4=I-4
    I5=I-5
    IC=I3
    D1E=(E(I5)-8.*(E(I4)-E(I2))-E(I1))/DR12
    D2E=(16.*(E(I4)+E(I2))-E(I5)-E(I1)-30.*E(I3))/DRS12
    DDE(IC)=D2E+D1E/R(IC)
2 CONTINUE

D1E=(3.*E(I1)+10.*E(I2)-18.*E(I3)+6.*E(I4)-E(I5))/DR12
D2E=(11.*E(I1)-20.*E(I2)+6.*E(I3)+4.*E(I4)-E(I5))/DRS12
DDE(I2)=D2E+D1E/R(I2)
D1E=(25.*E(I1)-48.*E(I2)+36.*E(I3)-16.*E(I4)+3.*E(I5))
1/DR12
D2E=(35.*E(I1)-104.*E(I2)+114.*E(I3)-56.*E(I4)+11.*
1E(I5))/DRS12
DDE(I1)=D2E+D1E/R(II)
DO 3 I=1,NR
D(I)=-(0.0,.5)*(DDE(I)+RK*(E(I)**2)*(DCONJG(E(I))))/
1WAVNUM-ABS0*E(I)/2.
3 CONTINUE
RETURN
END

C---------------------------------------------------------------C
C INTERPOLATE THE MIDPOINTS
C---------------------------------------------------------------C
SUBROUTINE INTERP(DZ,RK,WAVNUM)
COMPLEX*16 EO (200) ,E1. (200) , E2 (200) ,E3 (200) , D1 (200) ,
1D2(200), D3(200)
COMPLEX*16 RK,E1HAF (200) ,E3HAF (200) ,PC(200)
REAL*8 R(200),DR,Z,DZ,WAVNUM
REAL*8 A1,A2,B3,B6,A3,A9,DZ15,DZ90,DZ3,DZ54,DZ27
INTEGER*4 I,NR
COMMON/FIELDS/ EO,El,E1,E2,E3,D1,D2,D3,PC
COMMON/RADIOS/R,DR,NR
COMMON/DZFACT/ A1,A2,B3,B6,A3,A9,DZ15,DZ90,DZ3,DZ54,
1DZ27
DO 1 1=1,NR
E1HAF(I)=(80.*E3(I)+135.*E2(I)+40.*E1(I)+E0(I) +
1DZ15*(D1(I)-D3(I))+DZ90*D2(I))/256.
E3HAF(I)=(12.*E3(I)+135.*E2(I)+108.*E1(I)+E0(I)-
1DZ3*D3(I)-DZ54*D2(I)+DZ27*D1(I))/256.
1 CONTINUE
DO 2 1=1,NR
EO(I)=E3HAF(I)
E1(I)=E2(I)
E2(I)=E1HAF(I)
D1(I)=D2(I)
2 CONTINUE
CALL DIPEQU(E2,D2,DZ,RK,WAVNUM)
RETURN
END

C---------------------------------------------------------------C
C CALCULATE POWER
C---------------------------------------------------------------C
SUBROUTINE POWER(E,P,DZ)
COMPLEX*16 E(200)
REAL*8 R(200),DR,P,PO,ERE1,DET,PHS,COF,ABSPC,PI,ALP1,
1ALP2,DZ
INTEGER*4 I,NR,NR1,IOUT,IICHK
COMMON/BLOCK/ ERE1,DET,PHS,COF,IOUT,IICHK
COMMON/FACTOR/ ABSPC,ALP1,ALP2
COMMON/RADIOS/ R,DR,NR
PI=3.14159265359
NR1=NR-1
P0=P
P=P.0
DO 10 I=1,NR1
  P=P+PI*DR*(R(I)*CDABS(E(I))**2+R(I+1)*CDABS(E(I+1))
  1**2)/COF**2
10 CONTINUE
P=P*1.D-10
RETURN
END

C-----------------------------------------------C
C PROPAGATE TO THE DETACTOR POSITION C
C-----------------------------------------------C

SUBROUTINE FRENEL(E,E31,WNK,LAM,Z,RO)
INTEGER*4 NR,KK,NRK,IER,I,J,IOUT,IICHK
REAL*8 R(200),R1(200),DR,LAM,Z,RO,WNK,PI,MMBSJ0,ARG2
REAL*8 RMAX,ZR,ERE1,DET,PHS,COF,SPDL
COMPLEX*16 E(200),E31(200),FACTOR,ARG1,F1,F2,F3
COF=DSQRT(8.0*PI/SPDL)
KK= (NR-1)/2
KK=NR-1-2*KK
NRK=NR-1-KK
ZR=PI*R0**2/LAM
RMAX=3.*DSQRT(R0**2*(1+Z**2/ZR**2) )
DO 1 1=1,NR
  R1(I)=RMAX*(I-1)/NR
  FACTOR=-2.*PI*(0.,1.)/(LAM*Z)
  FACTOR=FACTOR* CDEXP((0.,1.)*WNK*Z *(l.+Rl(I)**2/(2*Z
  1**2)))
  E31(I)=(0.0,0.0)
  DO 2 J=1,NRK,2
    ARG1=(0.,1.)*WNK*R(J)**2/(2.*Z)
    ARG2=WNK*R1(I)*R(J)/Z
    ARG2=MMBSJ0(ARG2,IER)
    F1=R(J)*E(J)*CDEXP(ARG1)*ARG2
    ARG1=(0.,1.)*WNK*R(J+1)**2/(2.*Z)
    ARG2=WNK*R1(I)*R(J+1)/Z
    ARG2=MMBSJ0(ARG2,IER)
    F2=R(J+1)*E(J+1)*CDEXP(ARG1)*ARG2
    ARG1=(0.,1.)*WNK*R(J+2)**2/(2.*Z)
    ARG2=WNK*R1(I)*R(J+2)/Z
    ARG2=MMBSJ0(ARG2,IER)
    F3=R(J+2)*E(J+2)*CDEXP(ARG1)*ARG2
    E31(I)=E31(I)+DR*(F1+4.*F2+F3)/3.
```plaintext
2 CONTINUE
E31(I)=E31(I)*FACTOR
1 CONTINUE
RETURN
END

C------------------------------------------------------------ C
C CALCULATE CONSTANTS
C------------------------------------------------------------ C

SUBROUTINE CALCON(DR)
REAL*8 A112,B9,B1,B121,A242,A16,A13,DR,DR12,DRS12,DRS6
COMMON/CNSTNT/ A112,B9,B1,B121,A242,A16,A13,DR12,
1DRS12,DRS6
A112=112./121.
B9=9./8.
B1=1./8.
B121=9./121.
A242=242./27.
A16=1./6.
A13=1./3.
DRS6=6.*DR*DR
DR12=12.*DR
DRS12=12.*DR*DR
RETURN
END

C------------------------------------------------------------ C
C CALCULATE CONSTANTS
C------------------------------------------------------------ C

SUBROUTINE CALDZE(DZ)
REAL*8 A1,A2,B3,B6,A3,A9,DZ15,DZ90,DZ3,DZ54,DZ27,DZ
COMMON/DZFACT/ A1,A2,B3,B6,A3,A9,DZ15,DZ90,DZ3,DZ54,
1DZ27
A1=4.*DZ/3.
A2=8.*DZ/3.
B3=3.*DZ/8.
B6=2.*B3
A3=726.*DZ/216.
A9=3.*A3
DZ15=15.*DZ
DZ90=90.*DZ
DZ3=3.*DZ
DZ54=54.*DZ
DZ27=27.*DZ
RETURN
END

C------------------------------------------------------------ C
C MEHRDAD MOHEBI
C------------------------------------------------------------ C
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