NBS TECHNICAL NOTE

449

Research on Laser Standards and Materials At the National Bureau of Standards

U.S. DEPARTMENT OF COMMERCE
National Bureau of Standards

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UNITED STATES DEPARTMENT OF COMMERCE
C. R. Smith, Secretary
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NBS TECHNICAL NOTE 449

ISSUED JUNE 1968

Research on Laser Standards and Materials at the National Bureau of Standards

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Price 30 cents.

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Research on Laser Standards and Materials at the National Bureau of Standards†

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A brief report is presented on research in the areas of laser standards and laser materials now in progress at the National Bureau of Standards. Included are the measurement of the power and energy output of lasers, their near and far field patterns and their optical properties. The physical and chemical properties of laser materials are studied in an attempt to relate such properties to the operating characteristics of lasers.

Key words: Holography, laser energy, laser materials, laser power, laser radiometry, laser standards, laser transitions, modulation of lasers, near and far field patterns, optical properties.

1. Introduction

With support from the Advanced Research Projects Agency, the National Bureau of Standards is carrying out a program of research on the development of standards for laser performance, and of research on laser materials. The standards aspect of the research is aimed at developing standard methods for the measurement of energy, power and near-and far-field patterns, as well as to provide a central facility for the calibration of energy measuring devices. The research on laser materials is concerned with their characterization both as optical elements (i.e., with problems of optical quality, etc.) and as materials (i.e., with problems of composition and other relevant physical properties). Work is, at present, concerned primarily with ruby and neodymium glass. Some work is also carried out on gases and semiconductor laser materials and on passive non-linear optical materials.

The reviews reported below are necessarily brief summaries addressed to the general reader.

For the benefit of readers who would like more detailed technical information, the reports generally include bibliographical references and relevant publications originating from NBS work. The latter being designated with an asterisk.

†The support for part of the work reported herein comes from three sources, namely, ARPA Materials Office, ARPA Project Defender, and Air Force Avionics Laboratory. In the report the funding source for a given project is indicated after the title by an (M) (Materials Office) or a (D) (for both Defender and AF Avionics Laboratory).

2. Laser Performance Evaluation

2.1. Laser Energy and Power Measurement (D)

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The objective of this task was to develop reliable measurement methods for determining the power and energy of pulsed lasers at wavelengths of 694.3 and 1060.0 nm and up to 100 J per pulse.

A system, permitting the comparison of our carefully calibrated liquid calorimeter[1] with other energy measuring devices has been set up, and facilities for such comparisons up to 100 J per pulse have been established.

We estimated that this comparison system has a precision of ± 2%.

Some of the progress during the past year (partially supported by NBS Project No. 2510150) consisted of:

(1) The accurate evaluation of low reflectivity beam splitters by means of interchangeable calorimeters. (This has increased the energy measurement range of pulsed ruby laser

*[1] Jennings, D.A., IEEE Trans. IM-15, 1966, p. 161.

energy from ≈ 10 J to ≈ 100 J.)

(2) The design and construction of two "optical waveguide" calorimeters for ruby and Nd glass laser energy output measurements from ≈ 1 to ≈ 50 joules.

(3) The construction and evaluation of a hollow gold and silver ball calorimeter. This device was found to be unsatisfactory because of the large rates of heat losses from the spherical surfaces.

(4) The evaluation, as a function of beam size and energy, of ten calorimeters from four companies and three Government agencies. This survey showed sources of disagreement.

(5) Partial evaluation of four additional NBS liquid cell calorimeters for thermodynamic properties and accuracy.

2.2. Laser Pulse Measurements (D)

D. McSparron

Metrology Division, Washington, D.C. 20234
Institute for Basic Standards

Since calorimetric techniques preclude the measurement of pulse shape, the work in this Division was concentrated on the development of standard radiometric techniques for measuring pulse shape and the energy in individual laser pulses by using fast phototube systems.[1] In this system, the laser flash is attenuated to a level which can be handled directly by the photo-emissive detector by diffuse scattering from MgO screens.

Cross-correlation of this technique with energy measurement by means of calorimetric devices called for the use of highly stabilized ruby and neodymium lasers systems. Such a system with 20 joule, 2 pps capability was purchased and checked out.

Since initial comparison of the radiometrically based phototube system[1] and the calorimetric techniques showed a discrepancy of some 9%, an improved calorimeter, having higher thermal stability, low mass and reduced heat losses was built.

Further comparison between the two techniques showed agreement to within 4%.

*[1] McSparron, D.A., Douglas, C.A., and Badger, H.L., N.B.S. Technical Note 418, November 1967.

2.3. C.W. Power Measurements (D)

D. McSparron

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Institute for Basic Standards

The direct applicability of conventional radiometric techniques to the measurement of CW laser power was confirmed experimentally with an 80 milliwatt He-Ne laser. A power meter consisting of a vacuum phototube, diffusing glass entrance window and 7.5 cm diameter MgO coated entrance sphere, was constructed. This meter was calibrated by 4 standard radiometric techniques:

(1) A filter radiometer calibrated with an NBS lamp standard of spectral irradiance. (2) A prism spectroradiometer similarly calibrated. (3) A radiation thermopile calibrated with an NBS lamp standard of total irradiance, and (4) an absolute radiometer calibrated by electrical heating. The results of the direct substitution calibrations of the power meter were as shown in Table I.

Table I

Calibrating Instrumentation	Laser Incident Power (milliwatts)	Power Meter Calibration (μA/W)
Filter Radiometer	39.4	2.080
Prism Spectroradiometer	40.1	2.045
Radiation Thermopile	82.5	2.095
Absolute Radiometer	31.1	2.068

Although this work involved a particular He-Ne laser, operated at a particular wavelength, 6328Å, the techniques used are applicable to a much larger class of calibration problems. Provided that the incident laser radiation is not so intense as to physically damage the optical components involved, the filter and prism spectroradiometers should be limited only by the wavelength region for which standards currently exist, 0.25 to 2.5 microns, and the radiation thermopile and the absolute radiometer should be limited only by the wavelength region for which their response can be assumed to be spectrally flat.

2.4. Extension of the Radiometric Techniques to Other Wavelengths (D)

D. McSparron

Metrology Division, Washington, D.C. 20234
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The principle of wavelength independence of black body absorbers makes this type of structure particularly attractive for radiation measurement.

Although several such black body thermopiles in the form of conical cavities incorporating absorptive losses in multiple reflections have been designed, they were particularly vulnerable to radiation damage.

In view of this, a modification of the technique, involving the attenuation of the laser beam by diffuse scattering from MgO screens prior to incidence onto a large area thermopile was developed.

Exposure of such a system, with a thermopile time constant of 3 seconds, to a laser source pulsed at 1 pps resulted in a "steady state" signal S_1.

By prior calibration against a standard source, S_1 may be related to a definite power incident on the thermopile. Since the rate of pulsing was known, the energy content per pulse could be determined.

Thus, if the pulse train reaching the thermopile was then terminated by a shutter and the thermopile allowed to return to equilibrium at room temperature, the exposure of the thermocouple to another single pulse resulted in a ballistic signal S_2 from the thermopile and this signal corresponded to the above energy per pulse.

Although this technique gave results which agreed to within about 3% of our phototube system, its application was limited to pulses whose shapes closely conformed to that used in the calibration of the device.

2.5. C.W. Laser Power Measurement (D)

D.A. Jennings

Radio Standards Laboratory, Boulder, Colorado 80302
Institute for Basic Standards

The objective of C.W. laser power measurement, (project 2510458) was to develop methods to measure C.W. laser power in the 1 to 10 watt range in the 400 to 1000 nm region of the spectrum, and to use these methods to compare other power measuring devices.

The initial approach was two-fold:

(1) To increase the power of the existing lasers (Ar and YAG-Nd) to the one watt level.

(2) To develop power measurement techniques using pulsed Ruby laser calorimeters (energy measuring devices) as integrators with timed shutters.

Measurements already made suggest that we should be able to achieve a ± 5% precision and accuracy.

We will be working very closely with Dr. Knecht at Wright-Patterson Field (Air Force) on this project.

2.6. CO_2 Laser Power Measurements (D)

L.J. Schoen

Atomic Physics Division, Washington, D.C. 20234
Institute for Basic Standards

Considerable consideration has been given to the design of a power measuring device for C.W. infrared lasers. Initial experiments will be carried out on two CO_2 lasers having power outputs in the range 5 to 75 watts. It is expected that when the final design is settled upon it will be possible to scale it up or otherwise modify the design so that measurements may be made on more powerful lasers.

Several tentative designs have been considered, but the final design has not yet been settled upon. We plan to use a calorimetric type of measurement based on some type of steady state differential temperature measurement. To this end there has been considerable consultation with various calorimetry experts here at the Bureau of Standards.

The calorimeter will be filled with an absorbing gas so that the device need not be constructed in the form of a blackbody. Preliminary experiments indicate that SF_6 is a suitable absorbing gas. Calibration of the device will involve passing a known electrical current through a heater coil of known resistance in such a manner as to duplicate the power received from the laser beam.

2.7. Laser Power and Energy Measuring Techniques (D)

N.N. Winogradoff

Instrumentation Division, Washington, D.C. 20234
Institute for Applied Technology

The objective of this work was to investigate the general phenomena of the interaction of intense light with matter from the point of view of using these interactions to provide alternative means for measuring pulse shape, power and energy in laser beams.

Approaches Used:

(1) Elemental silicon as an attenuator.

(2) Optical rectification in non-linear optical materials.

Results and Conclusions: (Reported in the above sequence)

(1) In principle the pulse shape, power and energy in a ruby laser flash can be observed and measured by conventional photomultipliers and other photo-emissive detectors if the incident beam is attenuated by a suitable filter or diffusing screen. Most of the filters and screens commonly used consist of glass or other chemical compounds, and, as such, are susceptible to bleaching. Since radiative recombination of excess carriers in an indirect band gap material is a very inefficient process, it seemed feasible to use silicon as a "wavelength converting" attenuator. The ruby radiation was absorbed on the front surface of a polished Si wafer 0.10 cm in thickness, mounted directly on the window of an S1 photomultiplier. The 1.1 μm recombination radiation emitted from the rear surface was then measured by the photo-current in the detector.

Although the Si used had a lifetime of 400 μ sec, the photomultiplier current clearly reproduced laser pulses down to 2×10^{-8} seconds[1] (the limit of the electronic equipment used).

This technique enabled us to display the details of the spikes of a ruby laser on an oscilloscope.

The chemical inertness, high melting point and elemental nature of the Si eliminate the bleaching problems found in compounds. Attempts to obtain quantitative data relating the dependence of the attenuation factor on the beam intensity and semi-conductor characteristics showed that tight control over the flash lamp voltage and rod temperature was essential.

A Korad ruby laser system with a 25 joule capability and voltage and temperature control of ± 1 volt in 5000 and ± 1/4°F respectively was ordered and is being installed.

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*[1] Winogradoff, N.N., and Kessler, H.K., - Radiative Recombination Lifetimes in Laser Excited Silicon, Appl. Phys. Letters 8, 99 (1966).

(2) Conversations with Dr. J. Ward at the Clarendon Laboratories, Oxford, confirmed our belief that the dc field produced in a suitable non-linear crystal during the generation of second harmonic radiation could be measured and used to measure the E field in the laser beam. Theoretical relationships between this dc field and the other electro-optical coefficients of the material would provide means for discriminating between the voltage due to the optical rectification and other spurious effects.

It is hoped to undertake this investigation when the above Korad laser has been installed.

2.8. Far and Near Field Studies (D)

M.M. Birky

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Institute for Basic Standards

Preliminary studies (under ARPA support) of near and far field patterns of ruby are being made as a means of evaluation of optical rod quality. These patterns give information on homogeneity of active material and on beam divergence.

The far field pattern was obtained by placing a photographic film at the focal plane of a lens as shown in Fig. 1. If the lens has a focal length f, the beam divergence of the laser is given by $\alpha = D/f$, where D is the diffracted spot size as recorded. In order to get a number of exposures of different intensities for each laser shot, a plane-parallel glass prism was inserted between the film and lens, and inclined at a small angle to the optic axis of the laser and lens system (Fig. 1). The prism was used in a fashion similar to a Lummer-Gehrcke plate except in that it served only as an attenuator, and the thickness of the prism was such that there was no overlap of the beams. A number of beams of successively reduced intensity emerged from the two faces of the prism, the intensity ratio of successive beams being determined by the angle of incidence Φ and the index of refraction of the prism. For the far field pattern the photographic film was placed behind the prism at the focal plane of the lens, i.e. at 1f from the lens.

With this arrangement, there was the possibility of using light from either or both faces of the prism. If the lens of focal length f is placed a distance 2f from the output end of the laser, the far field pattern will be recorded at a distance of 3f from the laser, or as stated above, at the focal plane of the lens. However, at a distance 4f from the laser, or 2f from the lens, the laser head will be in focus, and the pattern recorded here on the film will be the near field pattern. So, with the prism in place, the light emerging from the two sides of the prism could be used to obtain near and far field patterns simultaneously. The near field patterns, also recorded at various intensities, give information on the homogeneity of the active rod, and the complexity of the mode structure.

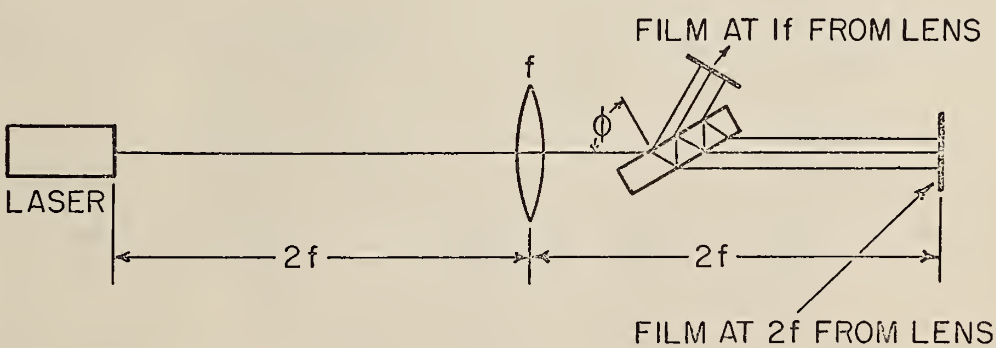

Figure 1. Arrangement for obtaining near and far field patterns generated by a laser

2.9. Mode Structure, Near and Far Field Patterns of CO_2 and Other Long Wavelength Lasers (D)

N.N. Winogradoff

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Objective: According to Capt. Axelrod of the Kirkland Air Force Base, there is an urgent need for a simple large aperture method of determining the above characteristics of CO_2 and other long wavelength lasers without cryogenic cooling of detectors.

Approach: At power levels in excess of a few milliwatts/cm^2, thermal quenching of a u.v. pumped phosphor provides a method for such observations.

Results and Conclusions: In collaboration with Dr. A. Ronn, and using a commercial phosphor, the feasibility and reasonable resolution of the method was demonstrated with a CO_2 laser.

A wide range of brilliant phosphors based on GeO_2 [1] and covering a wide spectral range have been found to be unimpaired after exposures to temperatures up to some $1200^{\circ}C$.

We plan to evaluate these phosphors for use in determining the above laser characteristics at higher power levels, and apply the same principles to ruby and Nd lasers.

[1] Marinace, J. and Winogradoff, N.N., GeO_2 Phosphors, Patent Applied for and assigned to I.B.M. (1960).

3. Materials

3.1. Bulk Optical Properties of Laser Materials (M)

G.W. Cleek

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Institute for Materials Research

This project was authorized in November 1967, with the objective of evaluating the bulk optical properties of laser crystals and glasses, particularly synthetic single crystal ruby and neodymium-doped glass. An analysis of the thermal distortion introduced into ruby and glass laser rods by flash lamps indicate that experimental data are needed on the change of refractive index with temperature, dn/dT, the change of refractive index with density, dn/dρ, and the thermal expansion, α. It is also necessary to know the elastic and photoelastic constants. The elastic constants of the glass will be determined by a pulse-echo technique, and the elastic constants of ruby are already known. It is planned to measure the remaining properties using a combination of interferometric and polarimetric techniques. Equipment is available to make appropriate measurements over the temperature range from -190° to $1000^{\circ}C$ and at pressures up to 1000 bars. In addition, the specimens will be examined for homogeneity and for inclusions.

A piece of Nd-doped glass in the form of a cylinder 8 cm in diameter and 2.9 cm thick has been supplied by the American Optical Company, as material for measurement of dn/dT, dn/dρ, α, elastic constants, photoelastic constants, density and refractive index. This specimen was marked for identification as 0835-AT. The two faces were polished and photographs were made of the fringe pattern in the Twyman-Green interferometer and of the shadowgraph pattern. These give an indication of the refractive index difference within the piece and of the optical homogeneity. The photographs are shown as Figures 1a and b, respectively.

The specimen was then examined microscopically for inclusions with side illumination against a dark background. Three inclusions that appeared to be metallic were found. Two seeds were also found in the specimen. A photomicrograph of the largest metallic inclusion is shown in Fig. 2.

When inspected between crossed polarizers, the specimen showed an optical path difference of about 15 m /cm at the edge. The birefringence was high only at the edge and was practically zero over a large area near the center. It was felt that the birefringence could be reduced by annealing, so the specimen was annealed by holding for 24 hours at its annealing temperature of $464^{\circ}C$. It was then cooled at the rate of $1^{\circ}C$/hr. to 350° and at $2.5^{\circ}C$/hr. to $220^{\circ}C$ at which point the furnace was cut off and allowed to cool to room temperature. The annealed specimen exhibited a maximum optical path difference of about 2 mμ/cm at the very edge, and was practically zero elsewhere.

The cylinder is now being cut to provide samples for the measurement of dn/dT, dn/dρ, α, photoelastic constants, elastic constants, density, and refractive index. The largest of the inclusions will be cut out and an attempt will be made to verify its composition by analysis.

Earlier, a sample of American Optical Company Nd-doped glass identified as C-835-E was obtained. Measurements of refractive index on a prism sample of this glass were made by Irving Malitson of NBS. The results are given in Table 1 and are plotted in Fig. 3. Over the temperature range from 10^{o} to 22^{o}C, he found that dn/dT was about $+3 \times 10^{-6}/^{o}$C.

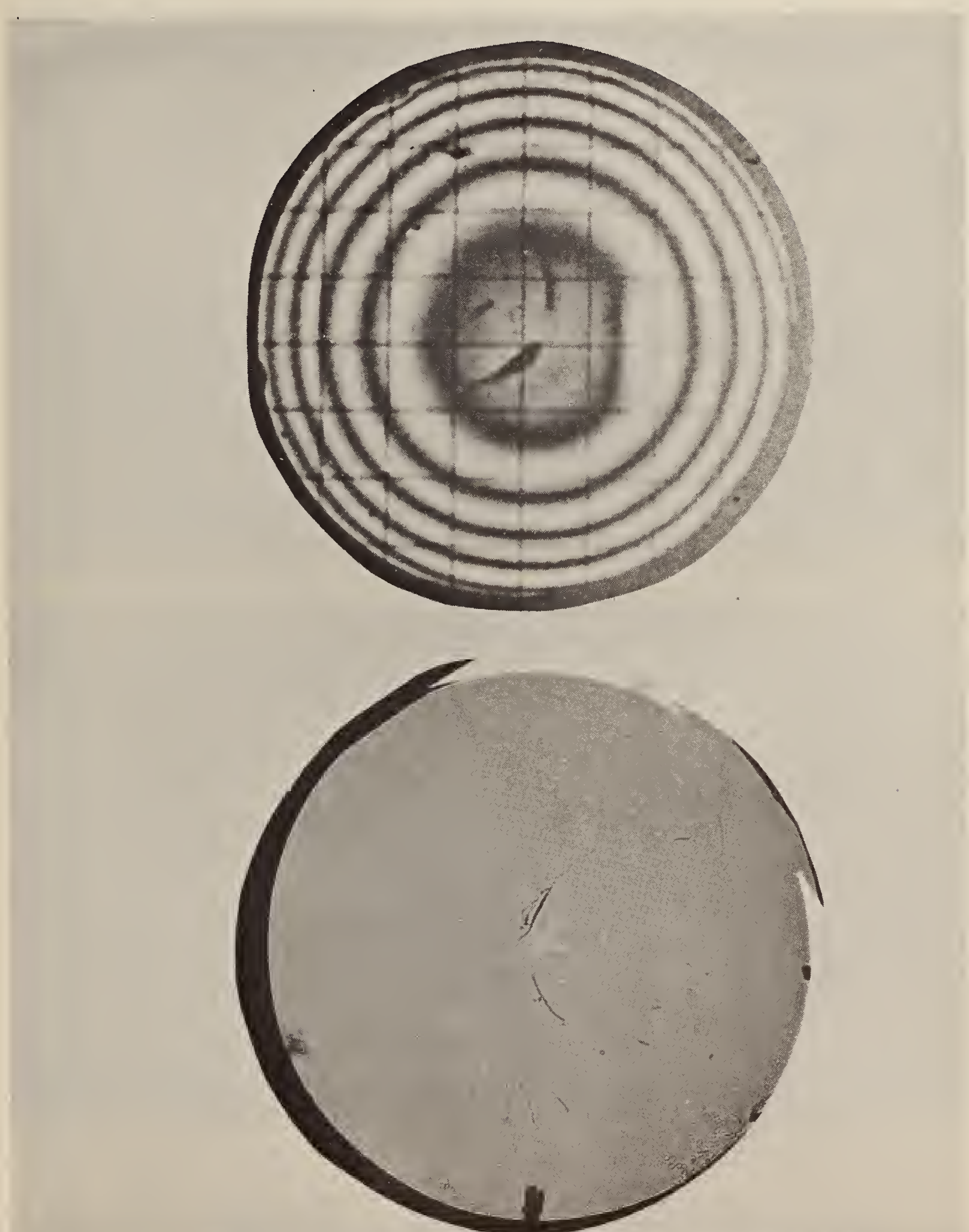

Figure 1a - Fringe pattern of specimen of glass 0835-AT photographed in Twyman-Green interferometer

Figure 1b - Shadowgraph photo of specimen of glass 0835-AT

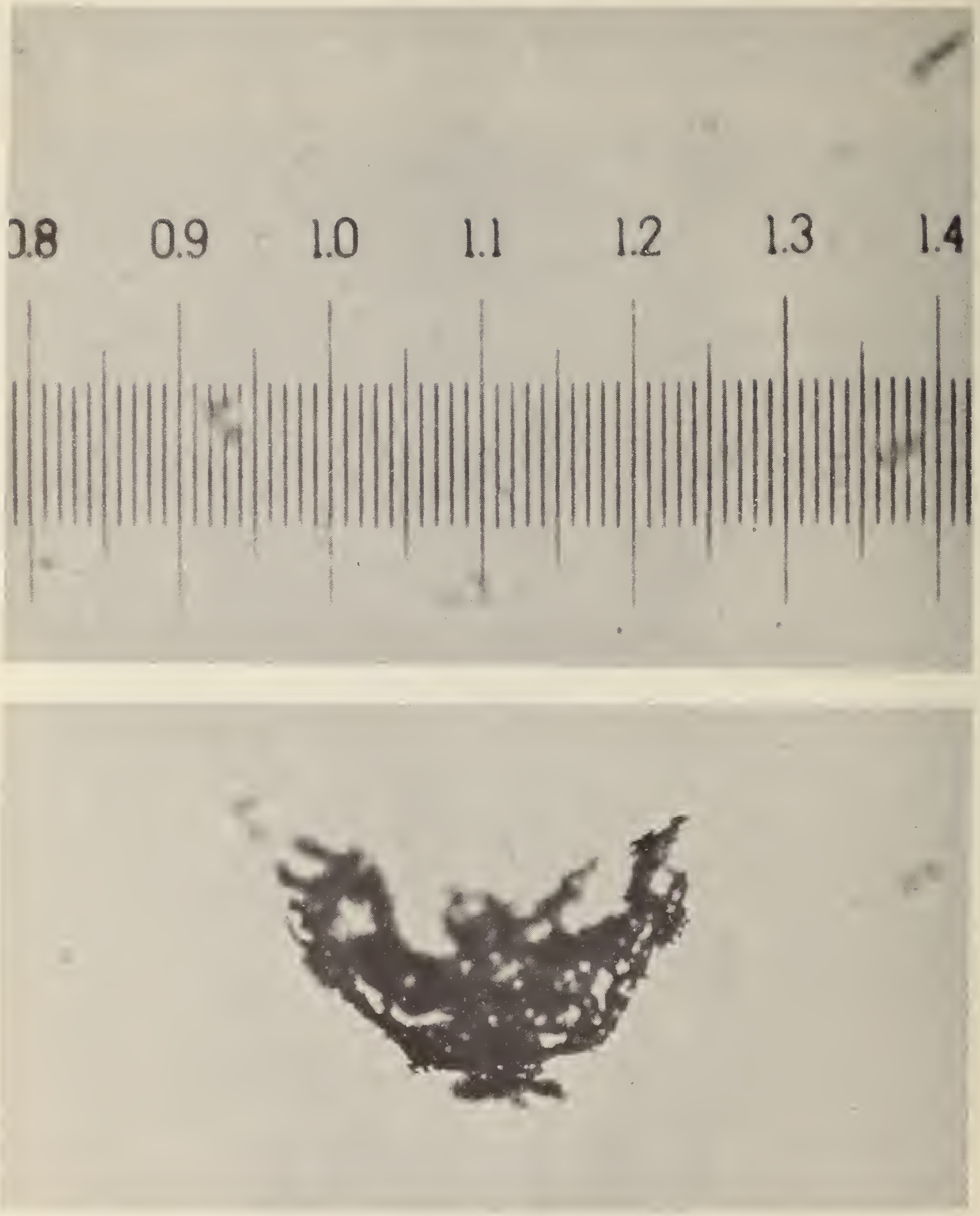

Figure 2 - Microphotograph of inclusion found in specimen of glass 0835-AT. A millimeter scale at the same magnification is shown at top for comparison.

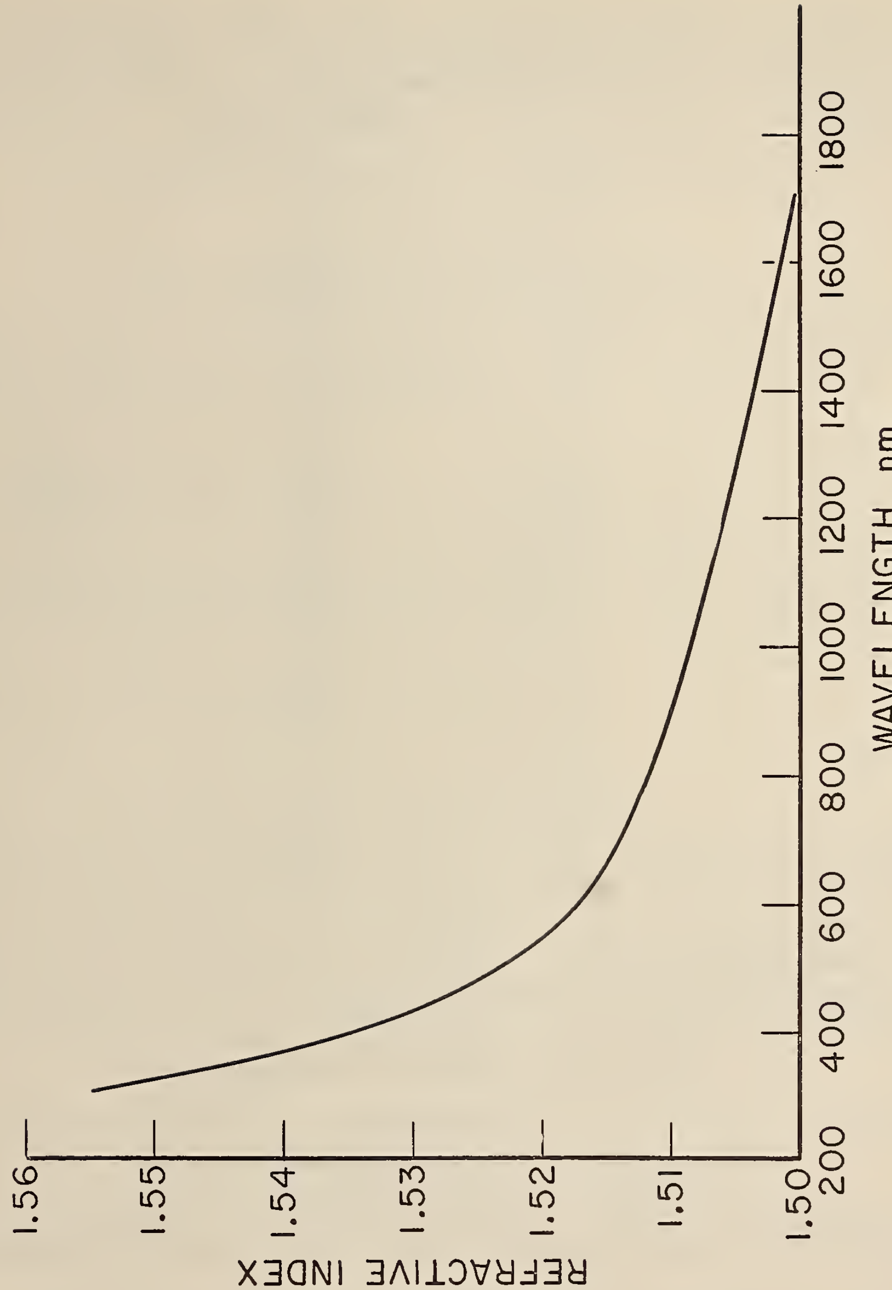

Figure 3 - Dispersion curve for glass C835-E

Table I

Refractive Index, Glass C-835-E
American Optical Co.

λ, nm	Refractive Index
313.0	1.55372
325.1	1.54938
340.4	1.54539
346.6	1.54375
365.1	1.53968
404.7	1.53295
435.8	1.52891
467.8	1.52566
480.0	1.52460
508.6	1.52242
546.1	1.52006
643.8	1.51574
667.8	1.51491
706.5	1.51377
794.7	1.51194
1014.0	1.50810
1083.0	1.50726
1128.7	1.50667
1362.8	1.50405
1529.5	1.50228
1709.1	1.50038

Uncertainty = $\pm 1 \times 10^{-5}$

dn/dT for 10^{o}C temperature range about $+3 \times 10^{-6}/^{o}$C

3.2. Characterization Standards for Laser Materials (M)

J.L. Torgesen

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This project was initiated in mid-November 1967, to develop precise methods for the chemical analysis of solid state laser materials and to provide characterization of laser materials as to physical defects, dislocations, etc.

Preliminary experiments in spark-source mass-spectrometry have been conducted by Mr. Paul Paulsen, Analytical Chemistry Division, NBS, with a sample of ruby of unknown source and a sample of neodymium glass. Mass spectrograms of good quality were obtained in both cases by employing (1) a high-purity gold counter-electrode directly and (2) repeating after sputtering a gold film on the sample surface in situ. No attempt has been made to interpret the mass spectrograms quantitatively in these trial experiments designed to explore methods of sparking with non-conducting materials.

Small disc specimens have been cut from ruby laser rods supplied by Mr. Mirarchi, Fort Monmouth. Both Verneuil-grown and Czochralski-pulled specimens are represented. Attempts are in progress (1) to spark specimen pairs directly; (2) to provide comparative analyses on specimens grown by different techniques; and (3) to establish the feasibility of using such pairs as reference specimens in the analysis of unknown samples in the spark-source mass spectrometer.

New ruby boules of high quality will shortly become available. These boules will contain chromium oxide dopant in nominal concentrations of 0.03, 0.05, and 0.07 weight percent. Specimens are to be taken from these boules for research on the chemical analysis of chromium with respect to its absolute concentration level in various oxidation states and its use as an internal standard for the determination of trace quantities of impurities. In addition, specimens are to be cut from the boules for the precise measurement of refractive index as dependent on chromium concentration and for the determination of stress-optical coefficients. These latter measurements are conducted under a related project.

3.3. Semiconductor Laser Materials and Laser Device Studies (M)

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Institute for Applied Technology

Scope - The work carried out was a continuation of the program described in NBS Report No. 9591 covering the period January 1, 1967 to July 1, 1967, and included:

1. A comparison of the characteristics of:

   (a) diffused
   (b) solution regrown, and
   (c) compensated, vapor deposited, epitaxial GaAs injection lasers.

2. The evaluation of GaAs lasers made by incorporating a wide range of dopings (Sn, Te and Zn) and degree of compensation of the p-type side of the junction with donors, by vapor phase epitaxy, to determine the optimum laser material specifications.

3. A study of the mechanisms responsible for the decrease in light power output when GaAs laser diodes are operated at other than cryogenic temperatures.

Results and Conclusions (Reported in the above sequence)

1. There has been considerable disagreement in the measurement of power levels of GaAs laser diodes when identical units were evaluated in different establishments.

Although this has been attributed to uncertainties in the calibration of the spectral response of the detector (usually a biplanar photo emissive cell) we have shown that in the pulsed mode, the differences observed were primarily due to the pulse shape of the current pulse generator, and that standardization of the rise and fall times of the current pulse, its duration and repetition rate is of primary importance in such comparisons. Slow rise times increased the temperature of the diode before an inverted population could be produced and resulted in low power output and high threshold currents. Similarly, when diffused laser diodes exhibiting a time lag between the onset of lasing action and the leading edge of the current pulse[1,2] were evaluated with short duration pulses, the production of an inverted population required very high peak currents, again giving the impression of high threshold currents.

Pulse generators producing 40 ns pulses with rise and fall times of $\simeq$ 4 ns at current levels of up to 140 A and having repetition rates of up to 400 pps have been constructed by H. Kessler and appear to be well suited to injection laser pumping.

2. Owing to difficulties in obtaining useable material, the evaluation of compensated, vapor grown epitaxial GaAs p-n junctions has been slower than expected. Since only 5 out of the 25 samples planned for, were obtained, no definite conclusions could be drawn so far.

3. The investigation of the temperature dependence of radiative recombination in GaAs by means of the Radiative Band Pinch Effect[3] showed that under ruby laser excitation, GaAs emitted radiation centered at 0.900 μm and that this radiation had a width at half height of 0.007 eV at room temperature.

Discussion of the experiment with Dr. Broom, of the University of Berne (Switzerland), lea the latter to suggest that this emission could be due to bound excitons.

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[1] Winogradoff, N.N., and Kessler, H.K., "Light Emission and Electrical Characteristics of Epitaxial GaAs Lasers and Tunnel Diodes," Solid State Commun. 2, 119, 1964.

*[2] Winogradoff, N.N., and Kessler, H.K., "Compensation and Band Tailing Effects in High Power Room Temperature GaAs Lasers" Solid State Commun. 5, 155, 1967.

*[3] Winogradoff, N.N., Owen, K., and Curnutt, R.M., "The Radiative Band Pinch Effect and Temperature Dependence of Radiative Recombination in GaAs" Int. J. Electronics 22, 229, 1967.

However, we could find no trace of such a line at low level injection but did observe a systematic shrinkage of the spectral width (observed by means of a thermocouple) as the ambient temperature was increased from 35°C to 230°C.

We have interpreted the new phenomenon of line narrowing with increasing temperature in terms of a temperature dependent shift of the Fermi level[4] in heavily doped material exhibiting exponential density of states tails in the conduction and valence band edges.[5] The experimental results again supported our contention that the decrease in power output of GaAs laser diodes at higher temperature was due to a decrease in the quantum efficiency for radiative recombination rather than to internal absorption.

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*4 Winogradoff, N.N., and Neill, A.H., Jr., "Band Tailing Effects and the Temperature Dependence of Radiative Recombination in Compensated Epitaxial GaAs Laser Materials," IEEE J. Quant. Electronics 1968 (in the press).

5 Pankove, J.I., "Absorption Edge of Impure Gallium Arsenide," Phys. Rev. A140, 2059, 1965.

*6 Winogradoff, N.N., "Field Control of the Quantum Efficiency of Radiative Recombination in Semiconductors," Phys. Rev. A138, 1562, 1965.

*7 Winogradoff, N.N., "Radiative Recombination within a Space-Charge Region in a Semiconductor," J. Appl. Phys. 37, 3916, 1966.

*8 Winogradoff, N.N., and Kessler, H.K., "Fermi Controlled Recombination as a Junction Design Factor in GaAs Laser Diodes," Int. J. Electronics 21, 329, 1966.

*9 Kessler, H.K., and Winogradoff, N.N., "Surface Aspects of the Thermal Degradation of GaAs p-n Junction Lasers and Tunnel Diodes," IEEE Trans E.D. ED13, 688, 1966.

3.4. Optical Evaluation of Laser Rods (D)

F. Rosberry

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Until recently, the program for the measurement of homogeneity in optical materials was primarily concerned with variations of the refractive index (measured to the 5th or 6th decimal place) in cylindrical samples measuring about 50 mm in diameter and 10 mm in thickness.

The results were reported in the form of contour maps with refractive index gradient lines enclosing areas of similar index as shown in Fig. 1.

The small frontal area and long length of the laser rods presented several problems in that small variations in refractive index tended to neutralize one another as the depth of the rod increased, while striae, inclusions, and voids increased with depth. Laser rods with beveled ends and end coatings made the optical evaluation of the rod material even more difficult.

In view of this, a procedure for evaluating the rod material by observing its transmission fringes and shadowgraph was developed. The interferograms were made on a Twyman-Green interferometer using a He-Ne gas laser source at 632.8 nm. The shadowgraphs were obtained by using a tungsten source in the apparatus shown schematically in Fig. 2.

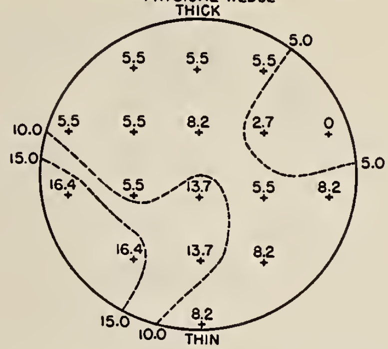
The overall quality found was somewhat below that of high quality optical glass due in part to the increased depth. Neodymium doped glasses and ruby crystal rods totaling eleven different samples have been examined to date. Figure 3 is a shadowgraph showing the striae present in a disk of neodymium glass. Windows were polished on the edge of this disk for refractometric measurements. Figure 4a shows a transmission shadowgraph through the length of a seven inch ruby crystal rod with beveled ends. Figure 4b is a reflection shadowgraph of a poorly finished end surface on a ruby rod. Figure 5a shows a transmission shadowgraph through about ten inches of a neodymium rod. The lower picture shows the interferogram of the same sample and presentation.

Four ruby crystal rods are now in our optical shop being refinished with ends perpendicular to rod axis in hopes of improving ease of transmission measurements.

VALUE FOR VARIATION OF INDEX OF REFRACTION

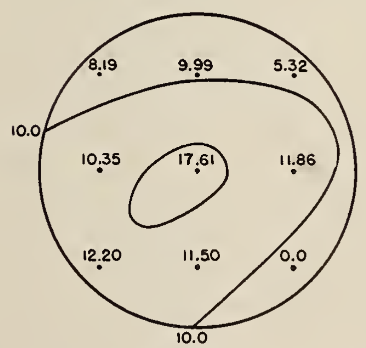
$\Delta n \times 10^{-6}$

PHYSICAL WEDGE
THICK



MAX. GRADIENT PER CM. = 10.9×10^{-6}
TOTAL GRADIENT 16.4×10^{-6}

SILICON
SINGLE CRYSTAL 1:1:1
"P" TYPE
$\Delta n \times 10^{-5}$



MAX. GRADIENT PER CM = 12.4×10^{-5}
TOTAL GRADIENT 17.6×10^{-5}

Figure 1 - Diagrams illustrating the comparison of the refractive index contour maps (showing areas of similar refractive index) in (a) a sample of high quality optical glass and (b) a single crystal of p-type silicon

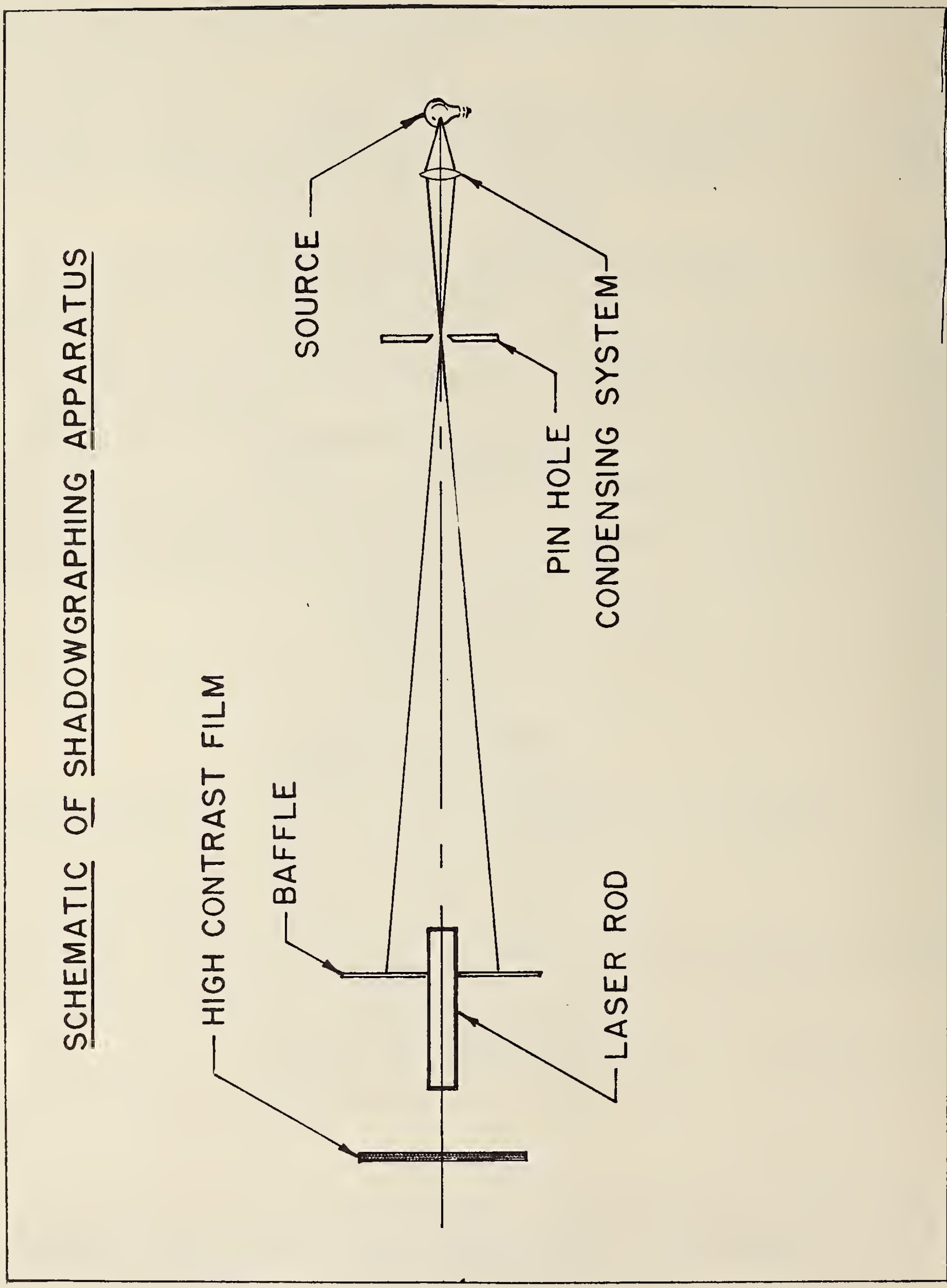


Figure 2 - Diagram of the shadowgraphing technique used in evaluating a laser rod

Figure 3 - Shadowgraph showing striae present in a disk of neodymium doped glass

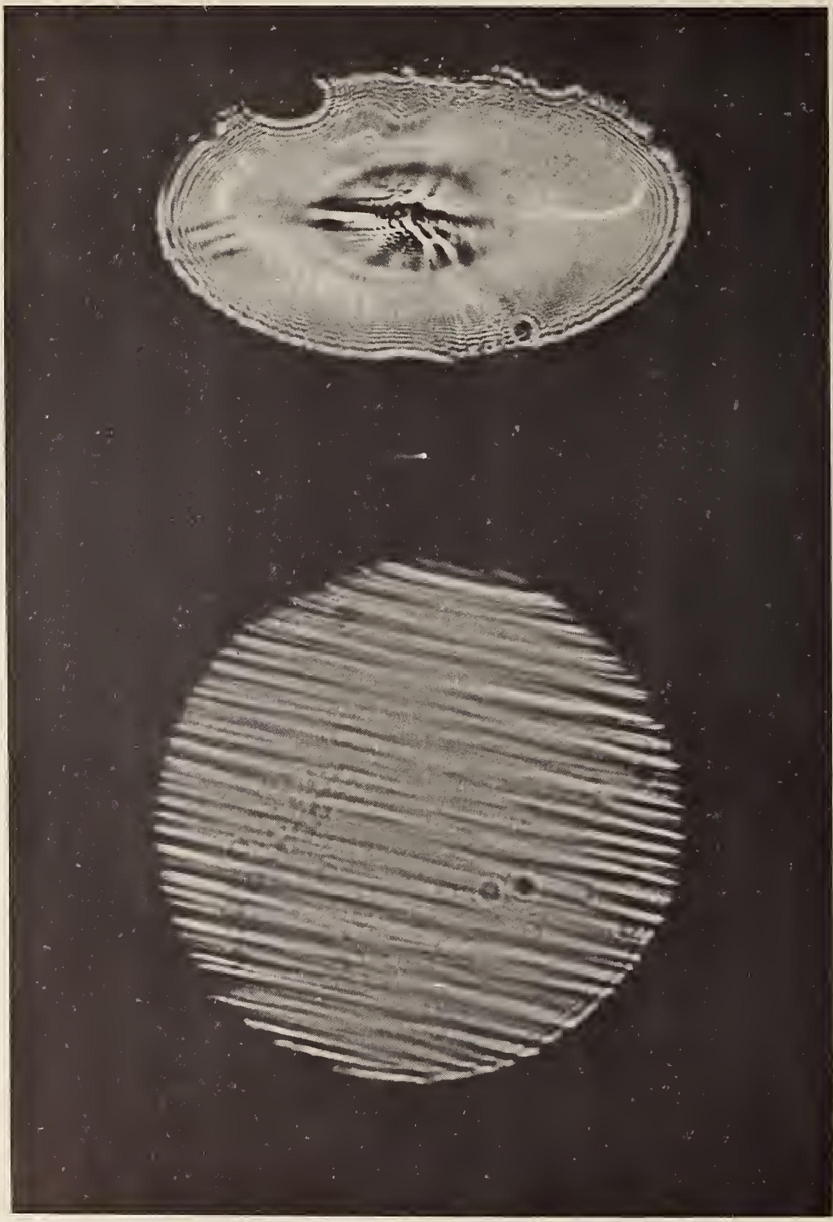


Figure 4a - Transmission shadowgraph through the length of a seven inch ruby crystal rod with beveled ends

Figure 4b - Reflection shadowgraph of a poorly finished end surface on a ruby rod

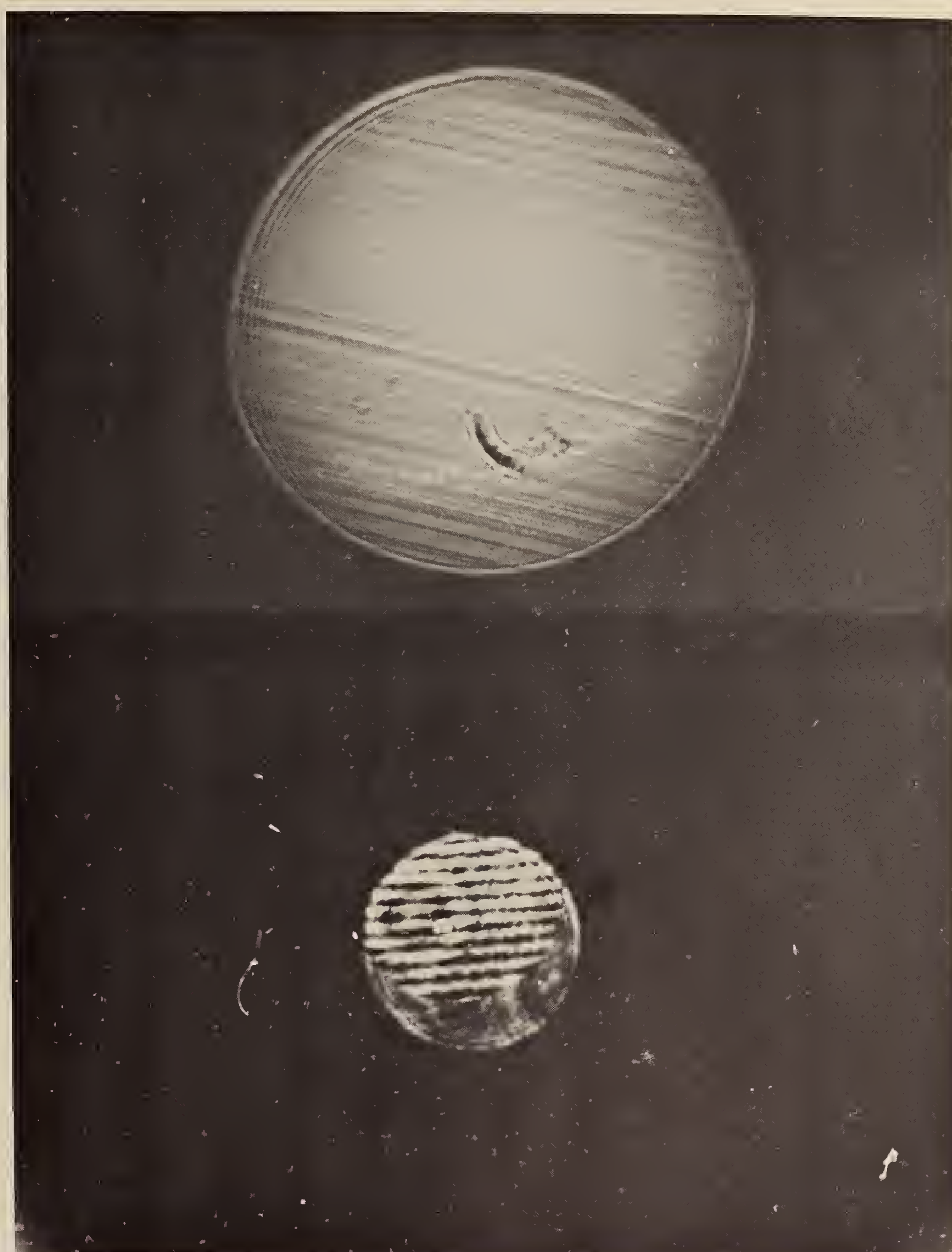


Figure 5a - Transmission shadowgraph through about ten inches of a neodymium doped glass rod

Figure 5b - Interferogram of the same sample

3.5. Magnetic Resonance Studies of Laser Materials (M)

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Since iron is present as an impurity in Nd glass laser rods, and the absorptive losses are governed by the relative amounts of iron in the Fe^{2+} and Fe^{3+} valency states, it was of interest to develop a technique for determining this ratio.

The approach used was to attempt to identify these ions by the fine structure of their paramagnetic resonance spectra. Preliminary results showed a somewhat asymmetrical line of about 2000 gauss in width at 4°K. It is hoped that heat treatment, partial crystallization or irradiation might produce a more distinctive spectrum which could be used for quantitative studies.

3.6. Laser Rod Holography (D)

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The various characteristics such as power and coherence of the light produced by a solid state laser are dependent on the laser rod makeup and the geometry of the rod and its cavity. It is of interest then to study the distortions introduced in a rod during pumping and lasing. In the past this has been difficult but currently the technique of holographic interferometry is enabling us to make such a study.

In this method, a hologram is made of the laser rod by shining light from a continuous laser through it and recording the interference between this light and light which has taken a path outside. When the hologram is returned to the position in which it was taken, interference occurs between the laser rod and its reconstructed image. If the arrangement has not changed in any way, no fringes will be formed. However, any changes made in the laser rod subsequently will show up as interference fringes. By the shape, spacing, and orientation of the fringes, the exact nature of the change can be determined.

To date, relaxation of distortions caused by heating a laser rod with a heat lamp and with pumping it have been observed. These measurements have demonstrated the basic sensitivity and accuracy of the method. Future plans currently call for increased time resolution so that observations can be made during the actual pumping and lasing conditions.

3.7. Crystal Characterization Studies by Optical Means (M)

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During this period the study of optical properties of $SrTiO_3$ in the region of the absorption edge has been completed. $SrTiO_3$ is an excellent prototype of the perovskite system. It lends itself well to many types of measurement (including transport properties, superconductivity, as well as the optical properties discussed here). The calculated band structure of Kahn and Leyendecker[1] serves as a most important "cornerstone" upon which meaningful investigations can be based. In this work absorption coefficient, reflectivity, and electro-reflectance measurements were made. The absorption coefficient shows an exponential rise (Urbach rule) over 5 orders of magnitude with a slope of 1/kT. The absorption data indicate a band gap of about 3.4 eV from the Urbach rule. This rule, simply stated as

$$\alpha = \alpha_o \, e^{\frac{\sigma (h\nu - E_g)}{kT}} ,$$

gives a cross-over of the exponential edges for all temperatures at $h\nu = E_g$. Here α is the absorption coefficient, $h\nu$ is the incident photon energy, and σ is a constant. Figure 1 shows the absorption edge at four temperatures ranging from 300°K on the left to near liquid helium temperature on the right. The extrapolated edges cross at $\alpha_o = 1.3 \times 10^{-4}$ cm^{-1} and 3.37 eV.

Both the D-C reflectivity and the electroreflectance spectra showed broad bumps near 3.4 eV in support of a 3.4 eV band gap.

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*[1] Kahn, A.H., and A.J. Leyendecker, Phys. Rev. 135, A1321 (1964).

Additional results on a heavily reduced sample show that Redfield's model[2] for an exponential edge is not applicable to $SrTiO_3$. This model (which would be appropriate for covalent semiconductors) requires freeze-out of carriers in order to yield the observed temperature dependent slope of an exponential edge. The reduced sample we measured, however, did not exhibit carrier freeze-out[3] and yielded the same temperature dependence as the pure material.

The electroreflectance spectra revealed several interesting features. The signal was quite large, extremely broad in energy, and of the wrong "sign," hence makes an identification with the Franz-Keldysh effect difficult. More typical F.K. data is presented by Shaklee, Pollak, and Cardona,[4] while Frova and Boddy[5] give electroreflectance spectra on several other "perovskite" crystals that resemble, in many respects, our $SrTiO_3$ data. The electroreflectance signal was appreciable down to at least 2.5 eV, well into the "window" of $SrTiO_3$. We thus feel that the electroreflectance effect observed could be due to a shift or a splitting in the major oscillator (4.4 eV),[6] responsible for the refractive index, with induced lattice polarization.

The optical properties of several other materials are being studied also. The solids are primarily high-symmetry crystals with high melting points and high dielectric constants. Included are TiO_2; the tetragonal scheelites, such as $PbMoO_4$; and "perovskites," such as $CsPbCl_3$ and $LiNbO_3$; the latter class being of considerable current interest since many exhibit "non-linear" optical behavior, and some have inordinately large electro-optical coefficients.

The room temperature absorption edges of several crystals are given in Fig. 2, showing the exponential behavior of the edge and the large anisotropy of $PbMoO_4$. This figure is a plot of log α (cm^{-1}) vs photon energy (eV).

To the above list we have recently added several crystals of the monoclinic $NiWO_4$ type. Some of these are known to be magnetic with very low Neel temperatures. Examples currently include $ZnWO_4$ and $MnWO_4$. The former appears to have a fundamental edge near 4 eV, while the latter (at least for crystals currently available) has a strongly anisotropic absorption band at about 2.2 eV.

In the past several months we have nearly completed the setting up of infrared equipment appropriate to a detailed comparative study of the "blue-band" that appears in reduced (or suitably doped) TiO_2, $SrTiO_3$, and Ba TiO_3. Measurements will involve optical absorption at various temperatures, and ultimately, photoconductivity. These will be correlated with transport measurements.

A paper, "Optical Properties of $SrTiO_3$ in the Region of the Absorption Edge," by M.I. Cohen and R.F. Blunt, is to be published in an April 1968 issue of The Physical Review.

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[2] D. Redfield, Phys. Rev. 130, 916 (1963).

*[3] H.P.R. Frederikse, W.R. Thurber, and W.R. Hosler, Phys. Rev. 134, A442 (1964).

[4] K.L. Shaklee, F.H. Pollak, and M. Cardona, Phys. Rev. Letters 15, 883 (1965).

[5] A. Frova and P.J. Boddy, Phys. Rev. 153, 606 (1967).

[6] M. Cardona, Phys. Rev. 140, A651 (1965).

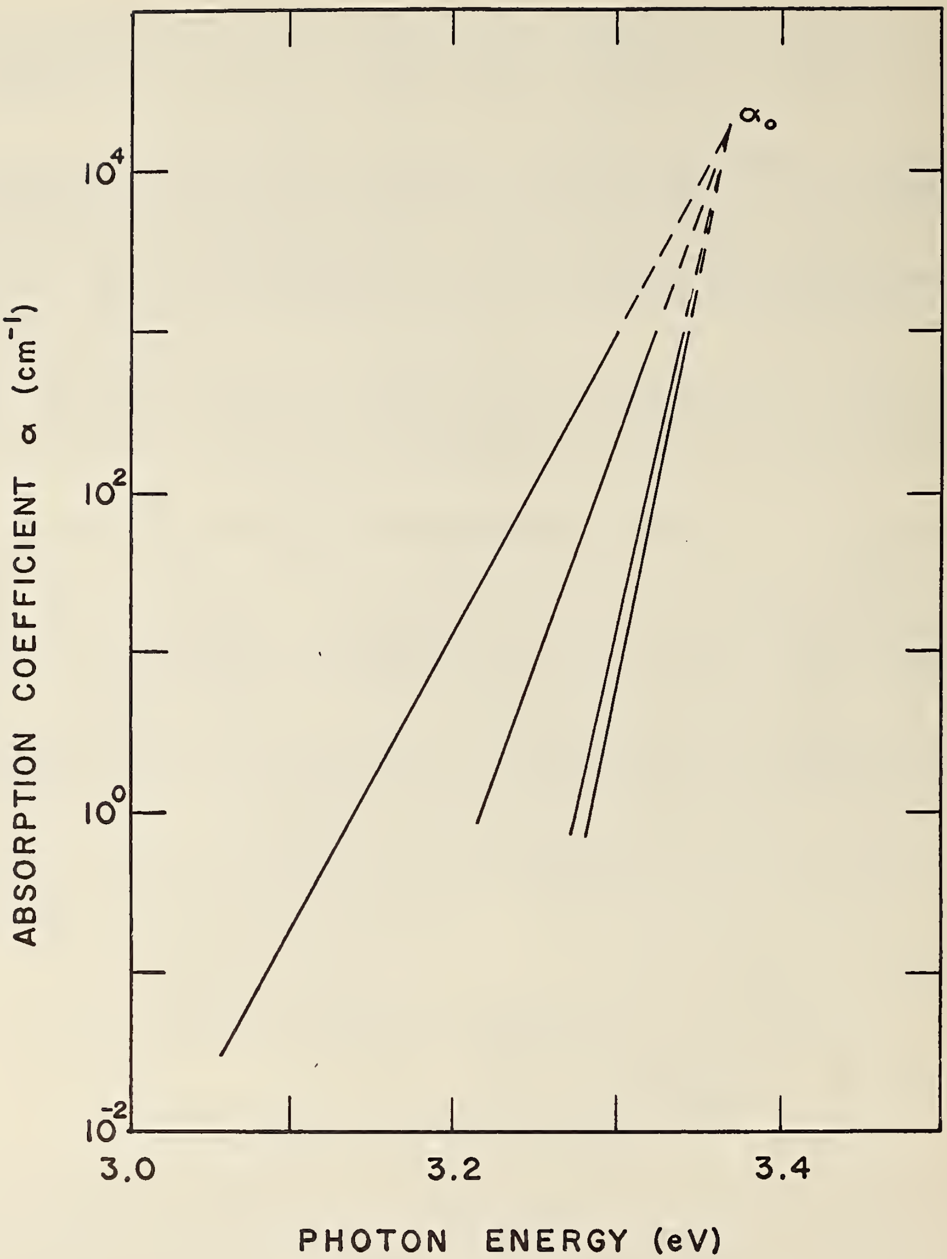

Figure 1 - Diagram illustrating the temperature dependence of the absorption edge of $SrTiO_3$

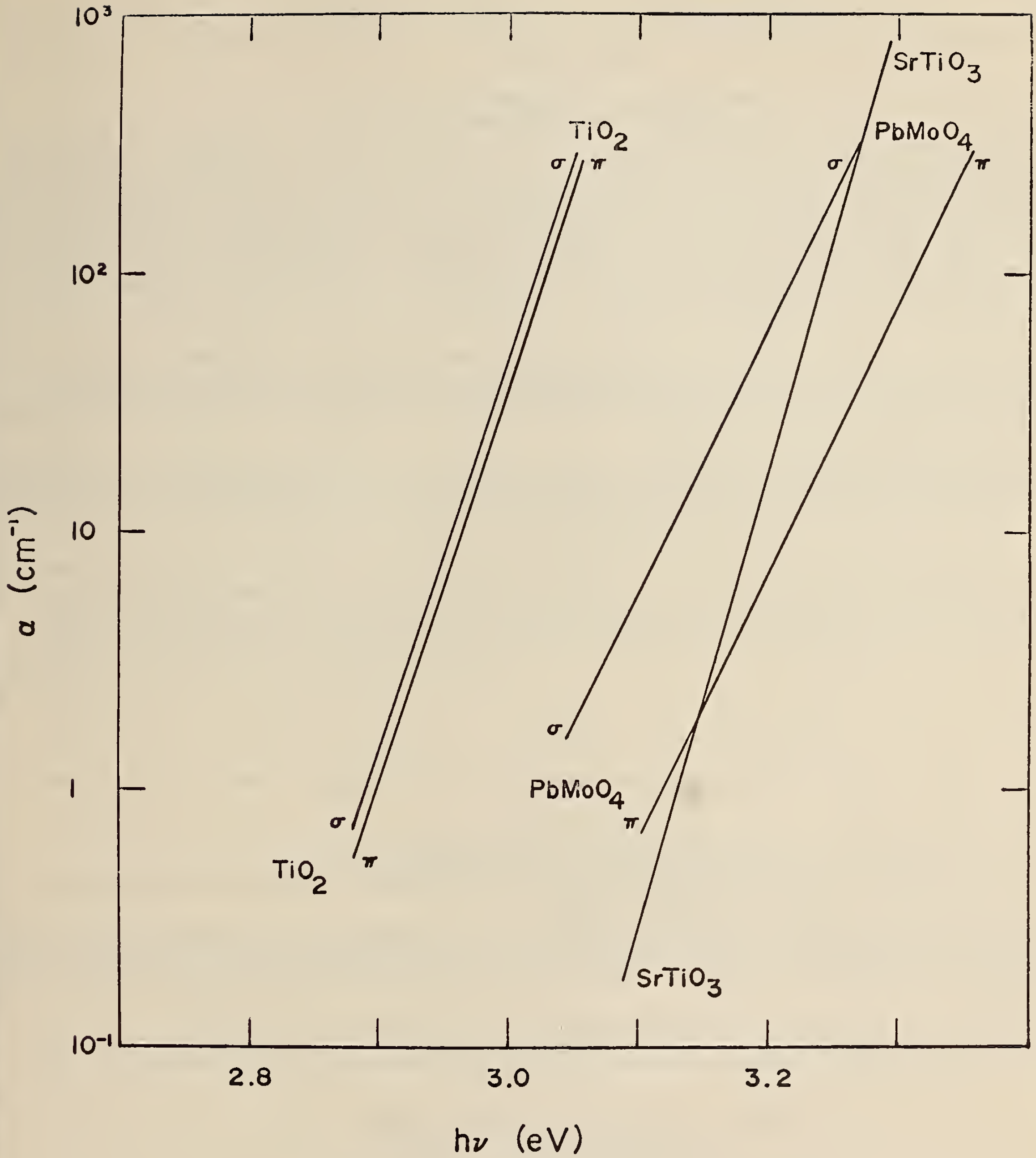

Figure 2 - Diagram showing the exponential nature of the absorption edges of TiO_2, $SrTiO_3$ and $PbMoO_4$ at room temperature

3.8. Liquid Lasers-Light Scattering in Liquids (D)

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The frequency distribution of initially monochromatic light which is scattered by density or composition fluctuations in a dense fluid is directly proportional to the frequency spectrum of those fluctuations. The frequency spectrum of the fluctuations can be calculated in terms of the material properties (specific heat, viscosity, etc.) of the fluid using the linearized hydrodynamic equations of irreversible thermodynamics. Such calculations have been carried out for fluids with internal degrees of freedom which are weakly coupled to the translational degrees of freedom. These calculations provide criteria for making a distinction between frequency dependent transport coefficients and additional thermodynamic variables which are not in local thermodynamic equilibrium.

Various aspects of critical opalescence in binary mixtures have been studied. The late Peter Debye and coworkers studied the shift in the critical mixing temperature when an electric field is applied and were able to observe the approach to the new equilibrium state of the fluid. We have developed a consistent means of relating these measurements to measurements of the frequency spectrum of light scattered by concentration fluctuations in the fluid.

The light scattering measurements presently conducted by Dr. Wims in collaboration with Dr. Sengers are aimed at studying the behavior near the critical point of binary mixtures. Simultaneously the critical behavior of the coexistence curve and the surface tension are being studied.

Relaxation processes which are not due to the exchange of energy between vibrational and translational degrees of freedom have been observed in many liquids. These are called "structural relaxation" processes as it is assumed that the relaxation mechanism involves changes in the local order of the fluid in response to fluctuations in the density "Structural relaxation" has been observed in CO_2 near the critical point. An effort is being made to characterize this effect in terms of correlation functions.

[*1] Mountain, R.D. and T.A. Litovitz, "Negative Dispersion and Brillouin Scattering," J.O.S.A. 42, 516 (1967).

[*2] Mountain, R.D., "Density Fluctuations in Fluids Having an Internal Degree of Freedom," J. Res. NBS 72A, 95 (1968).

[*3] Mountain, R.D. and Cooper, M.J., "Interpretation of Relaxation Times in the 'Electric Field Effect on Critical Opalescence'," J. Chem. Phys. (1968) (to be published).

[*4] Larsen, S.Y., Mountain, R.D. and Zwanzig, R., "On the Validity of the Lorentz-Lorenz Equation Near the Critical Point," J. Chem. Phys. 42, 2187 (1965).

[*5] "Spectral Structure of Critical Opalescence: Binary Mixture," J. Res. NBS 69A, 523 (1965).

[*6] Mountain, R.D., "Spectral Distribution of Scattered Light in a Simple Fluid," Rev. Mod. Phys. 38, 205 (1966).

[*7] Mountain, R.D., "Interpretation of Brillouin Spectra," J. Chem. Phys. 44, 832 (1966).

[*8] Mountain, R.D., "Thermal Expansion and Brillouin Scattering in Liquids," J. Res. NBS 70A, 207 (1966).

3.9. Laser Transitions in HCN and DCN (D)

D.R. Lide, A.G. Maki and L.J. Schoen

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The so-called CN laser has been known for several years.[1] One commercial firm has been selling such lasers for some time. Yet, it was not until studies were initiated in the Infrared and Microwave Spectroscopy Section of the Bureau of Standards that it was shown that the HCN molecule is responsible for the laser action.[2]

Studies of the HCN and DCN laser systems are continuing in the hope that all known laser transitions can be identified. The work on the laser assignments for HCN and DCN has resulted in several successful

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[1] Gebbie, H.A., Stone, N.W.B. and Findlay, F.D., Nature 202, 685 (1964).

[*2] Lide, D.R. and Maki, A.G., Appl. Phys. Letters 11, 62 (1967).

attempts to find new laser transitions on the basis of theoretical predictions. The Bureau of Standards group has been in contact with Professor Javan at M.I.T. and his group has been able to verify the assignments by finding new laser lines at frequencies predicted by the Bureau's assignments.[3,4,5]

It seems that laser lines for both HCN and DCN are due to an unusually high population of molecules in certain vibrational levels. Where those levels come close to other unpopulated levels, laser action is possible as the energy in the populated levels is so to speak dumped into the unpopulated levels. This dumping can take place only if the transition is rather strongly allowed. In most cases the transitions are too weak for laser action. In a few cases the levels are connected through a Coriolis interaction. That is to say, the vibrational and rotational motion of the molecule gets mixed-up in such a way that the two vibrations resemble each other and, if they are close together, the molecule can hardly tell in which vibrational state it belongs. Under those circumstances the molecule can easily pass from one vibrational state into the other. That means that the transition between the two states is strongly allowed and laser action can result. This explanation holds for both HCN and DCN although the energy levels involved are different for the two isotopes.

The Infrared and Microwave Spectroscopy Section is continuing its study of the rotational and vibrational energy levels of HCN in an effort to provide more positive assignments for the remaining laser lines. In the past three months new infrared spectra have been obtained which verify some of the laser assignments and which make it possible to predict some more new and as yet unobserved laser lines.

A secondary result of the explanation of the HCN and DCN lasers was the fact that it demonstrated a new mechanism for explaining laser action. As a result, other workers were then able to explain the laser transitions observed in H_2O and D_2O lasers. That explanation had also eluded workers for several years although many laboratories throughout the world were studying such lasers.

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[3] Hocker, L.O. and Javan, A., Phys. Letters 25A, 489 (1967).

[4] Hocker, L.O. and Javan, A., Appl. Phys. Letters 12, 124 (1968).

*[5] Maki, A.G., Appl. Phys. Letters 12, 122 (1968).

4. General

4.1. HCN Laser Frequency Measurement (D)

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The objective of this task was to measure the frequency of the HCN laser. This has been done by beating a 74 GHz klystron with the laser output in a silicon diode. The frequency of the two prominent laser lines were found to be 890,759 ± 0.1 MHz and 964,312.1 ± 0.2 MHz.

4.2. Giant Pulse Mode Control (D)

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Radio Standards Laboratory, Boulder, Colorado 80302
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Experiments for the control of modes in giant pulsed lasers are being performed.

It is expected that axial mode control will be accomplished by using an etalon as an output mirror, non-axial modes will be controlled by one of two methods: (a) an aperture in the cavity which will restrict operation to the fundamental mode or (b) the use of optical components to force the entire rod to operate in the fundamental mode. When the laser is operative applications will be tried.

This project was started in November 1967; however, a new water cooled laser head has been constructed with emphasis on temperature control and uniform pumping of the laser rod. Preliminary investigations have been started of several types of etalons for mode control. A power monitor is also being constructed.

4.3. The Pressure Dependence of the $(3s_2\text{-}2p_4)$ 6328Å Transition in Neon (D)

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Atomic Physics Division, Washington, D.C. 20234
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The use of a stabilized laser for various types of length measurements requires a knowledge of its wavelength reproducibility whether it be used as a primary or secondary standard. One convenient means of stabilization that is commercially available is to lock the laser to the bottom of the "Lamb dip." How close this point is to the true atomic resonance frequency is dependent on the gas pressure and other effects. This investigation is limited to the Doppler broadened $(3s_2\text{-}2p_4)$ 6328Å ^{20}Ne transition.

Two lasers attached to a vacuum system with helium and neon reservoirs are being used to study the effects of gas pressure on the laser output frequency. Each laser consists of a 31 cm hemispherical cavity, 3 mm I.D. tube and is rf excited. The flat mirror is mounted on a piezoelectric transducer that is used for the length scan and the stabilization. They are independently stabilized on the bottom of the Lamb dips. The construction of the system allows the lasers to be filled to different pressures.

The frequency change as the pressure is increased in one laser is detected by looking at the beat between the two lasers, with a phototube, a spectrum analyzer, a counter and a digital recorder. The direction of the change is also recorded by continuously monitoring the error signal from the servo-system used to control the laser in which the pressure is changed. The correlation between the frequency shift and error signal voltage was verified initially with the aid of a Fabry-Perot interferometer.

Data taken between 1.2 to 3.2 torr show that the bottom of the Lamb dip increases in frequency with increasing ^3He pressure. The magnitude of the shift is about 17 MHz/torr in this pressure range.

The experimentally obtained Doppler broadened line with its Lamb dip superimposed has some asymmetry. The immediate problem is to separate any instrumental asymmetry (for example, mirror tilt during scan) from asymmetry in the atomic line, and to extend the measurements to lower pressures. Also, since all measurements by others have employed dc excitation of the laser, plans are to heterodyne a dc laser with an rf excited laser to determine if there is a Doppler shift as a result of a net drift of the radiating atoms.

4.4. Fluorescence and Double Resonance Phenomena in CO_2 (D)

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New laser induced infrared fluorescence experiments have been performed. These experiments consisted of studying the infrared emission spectrum of gases which were irradiated by a CO_2 laser. The laser raised the molecules from the ground vibrational state to some higher vibrational state. Fluorescence resulted as the molecules then emitted radiation in falling back to the ground vibrational state. These experiments should provide more information on both rotational and vibrational relaxation times and on the selection rules which govern collisional relaxation mechanisms. So far all experimental attempts to make quantitative measurements have been unsuccessful due to equipment difficulties. Some of this work is being done in collaboration with Dr. George Flynn's group at Columbia University. Qualitative work has been done on about ten compounds. Ammonia was the lightest molecule studied. It is potentially a very good molecule to use for these studies, because its rotational spectrum is fairly well understood. However, one really should have somewhat better resolution than is currently available to us in the 10 micron region.

Very little further work has been done on infrared and microwave double resonance experiments. In the last semi-annual report experiments were described where a CO_2 laser is used to vibrationally excite certain molecules while observing the effect on the microwave pure rotational spectrum. We hope to do some more double resonance work in the future.

[*1] Ronn, A.M. and Lide, D.R., Infrared and Microwave Double Resonance Using a CO_2 Laser, J. Chem. Phys. 47, 3669 (1967).

4.5. Crystal Defect Studies Using Magnetic Resonance (M)

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When ions of certain impurities are introduced into suitable crystals, the impurity ions interact with the surrounding host ions much in the same manner as the host ions interact among themselves.

However, the impurity ions will retain some of their own characteristics and may distort the local environment.

If the crystal is an insulator (or, in some cases, a conductor or semiconductor) and the impurity ions are paramagnetic (i.e. have some unpaired electronic spins), the method of electron paramagnetic resonance could be used to study the properties of both the host lattice and the impurities. In addition, if the energy states of the spin system between which the transitions were observed could be equally populated (i.e. the system is saturated) by pumping with relatively high intensity radiation, the relaxation processes toward thermal equilibrium may shed some light on the properties of either the host lattice, impurity ions, or both.

According to Van Vleck[1] the relaxation processes are temperature dependent. At very low temperatures, the spin system will relax by the direct emission of a phonon, i.e. by contributing to a certain mode of lattice vibration. The relaxation rate for this process is linearly dependent on temperature.

At higher temperatures where thermal lattice vibration is appreciable (i.e. the phonon spectrum extends into higher frequencies), the spin system can give up its energy to a phonon by collision, resulting in a phonon of higher energy. The relaxation rate for this two phonon or Raman process is proportional to the ninth power of the temperature.

At still higher temperatures, Orbach's mechanism[2] where the spin system is first raised to a higher (real) excited state by phonon absorption and then relaxes to the ground state with an exponential time dependence, dominates.

In general, if we denote the instantaneous population difference between two levels by n, and the population difference at thermal equilibrium by n_o, then

$$n = n_o \left[1 - \exp\left(- t/T_1\right)\right] \quad (1)$$

where t is time, and T_1 is a characteristic time called the "spin lattice relaxation time" which depends on both the ions, the host lattice and on the temperature at which the relaxation takes place.

The above temperature dependencies of the relaxation times $1/T_1$ for the various temperature ranges have been verified experimentally for many ions and particularly for ions of the rare earth elements.

Experiments with M_o^{5+} in TiO_2 [3] showed that at 4^oK saturation could easily be obtained with only a few milliwatts of microwave power, so that the study of the relaxation mechanism in this material required very low sampling power.

A high sensitivity, superheterodyne detection system was therefore constructed, and preliminary measurements with this system are shown in Figs. 1 and 2. The temperature dependence of the relaxation rates were found to be T^9 between 3^oK and 210^oK, linear below 1.5^oK, and T^5 between 1.5^oK and 3^oK. The latter is a new result and although this temperature dependence was reproduced under different experimental conditions and with different samples further experimental work to establish a true T^5 dependence is under way.

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[1] Van Vleck, J.H., Phys. Rev. 57, 426 (1940).

[2] Finn, C.B.P., Orbach, R. and Wolf, W.P., Proc. Phys. Soc. (London) 77, 261 (1961).

[3] Chang, T., Phys. Rev. 136, A1413 (1964).

[4] Yang, G.C. and Chang, T., Bull. Amer. Phys. Soc. 11, 220 (1966).

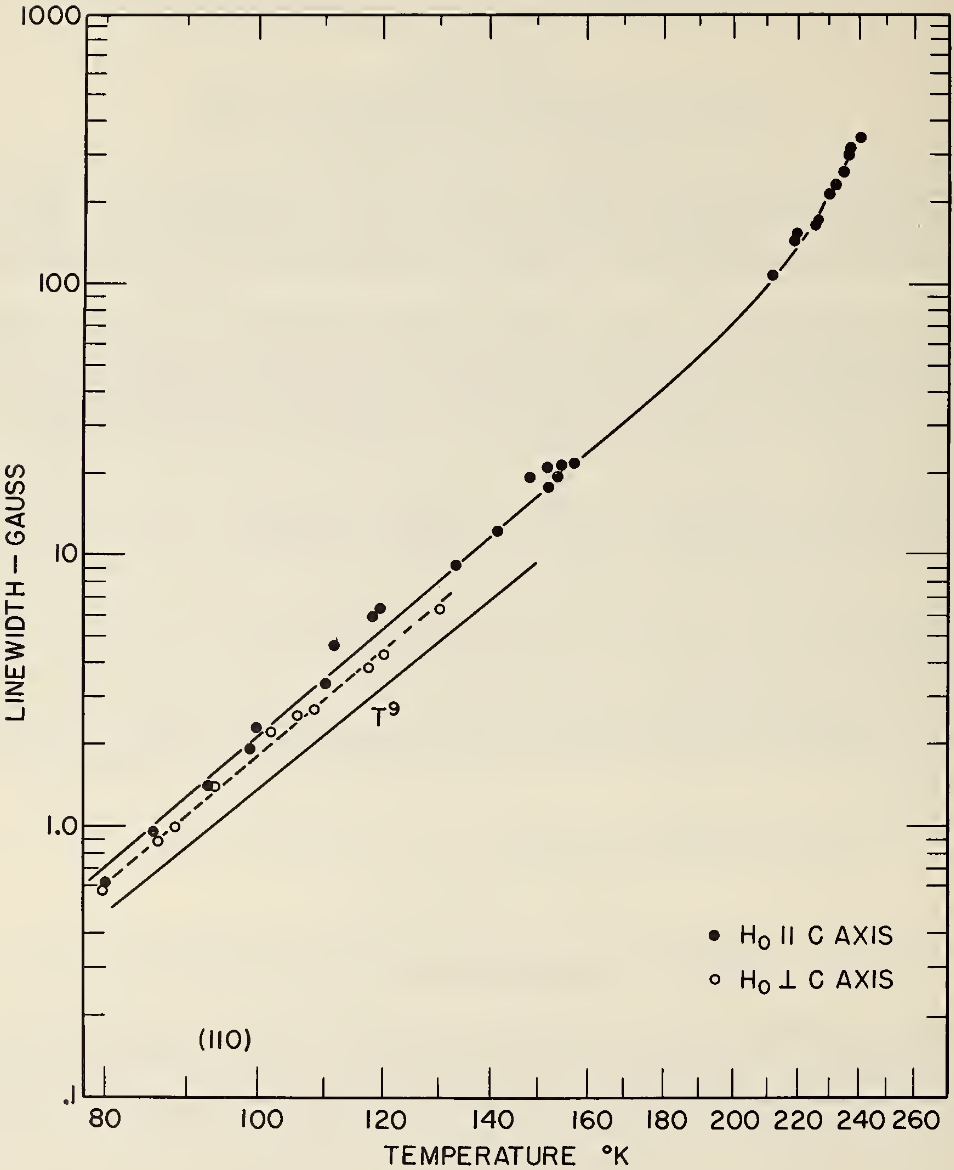

Figure 1 - The temperature dependence of the linewidth of $Mo^{5+}:TiO_2$. The circles and dots are experimental points. The line marked T^9 is a theoretical curve

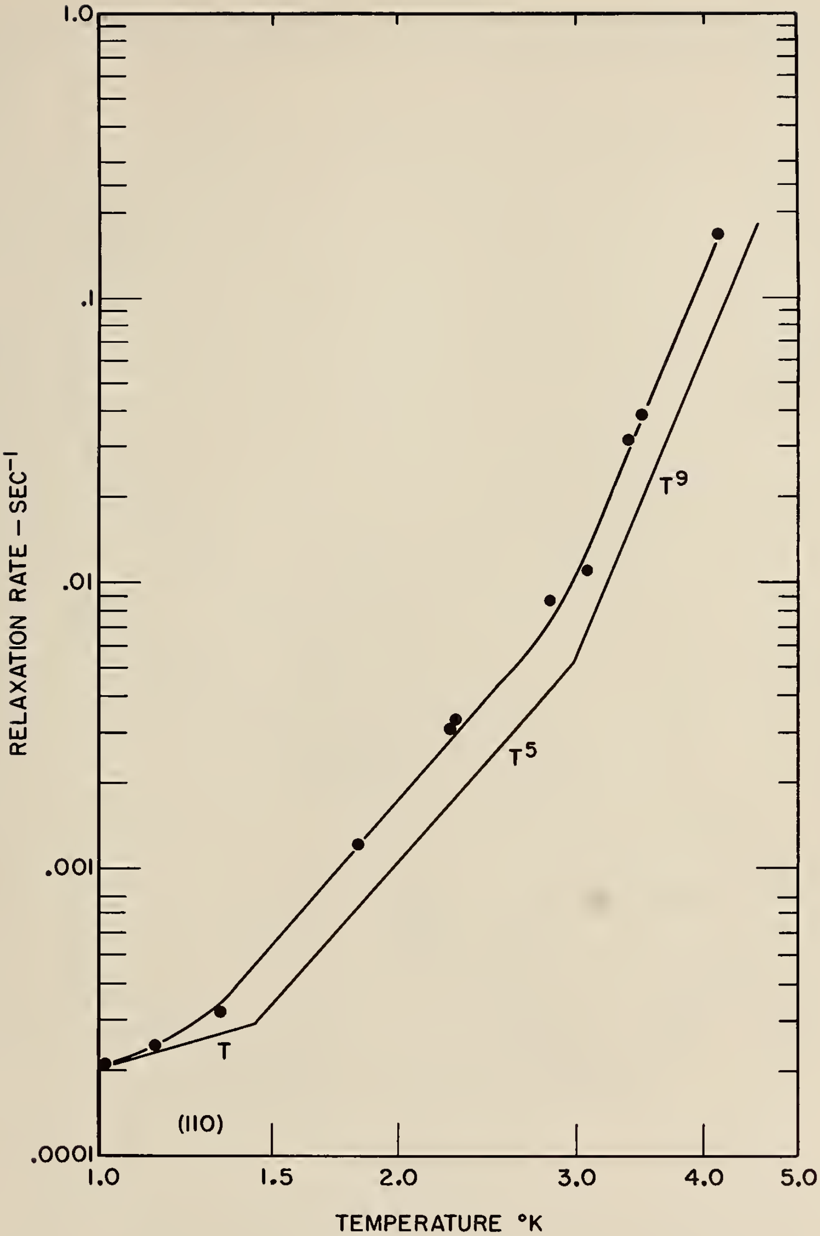

Figure 2 - The temperature dependence of the relaxation rate of $Mo^{5+}:TiO_2$. The dots are experimental points. The lines marked T, T^5, and T^9 are theoretical curves

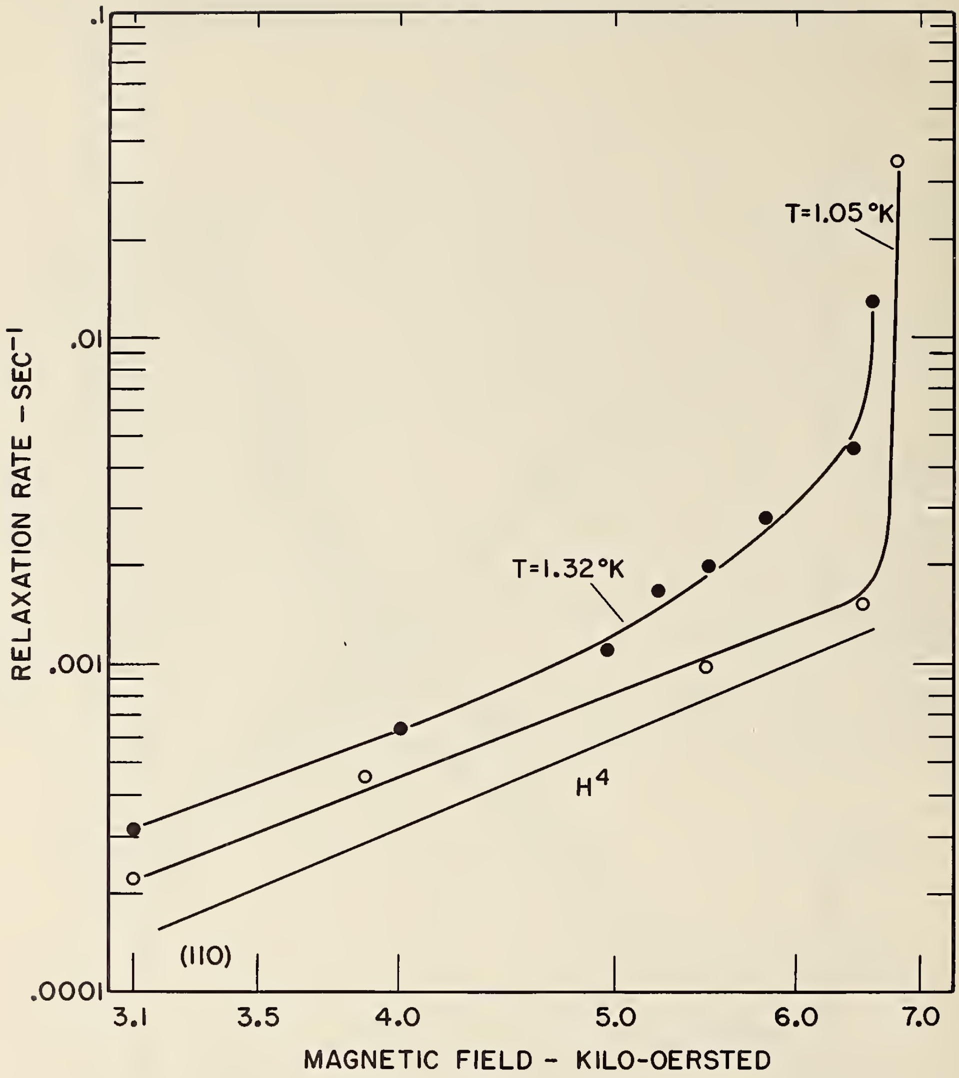

Figure 3 - The magnetic field dependence of the relaxation rate of $Mo^{5+}:TiO_2$. The dots and circles are experimental points. The line marked H^4 is a theoretical curve

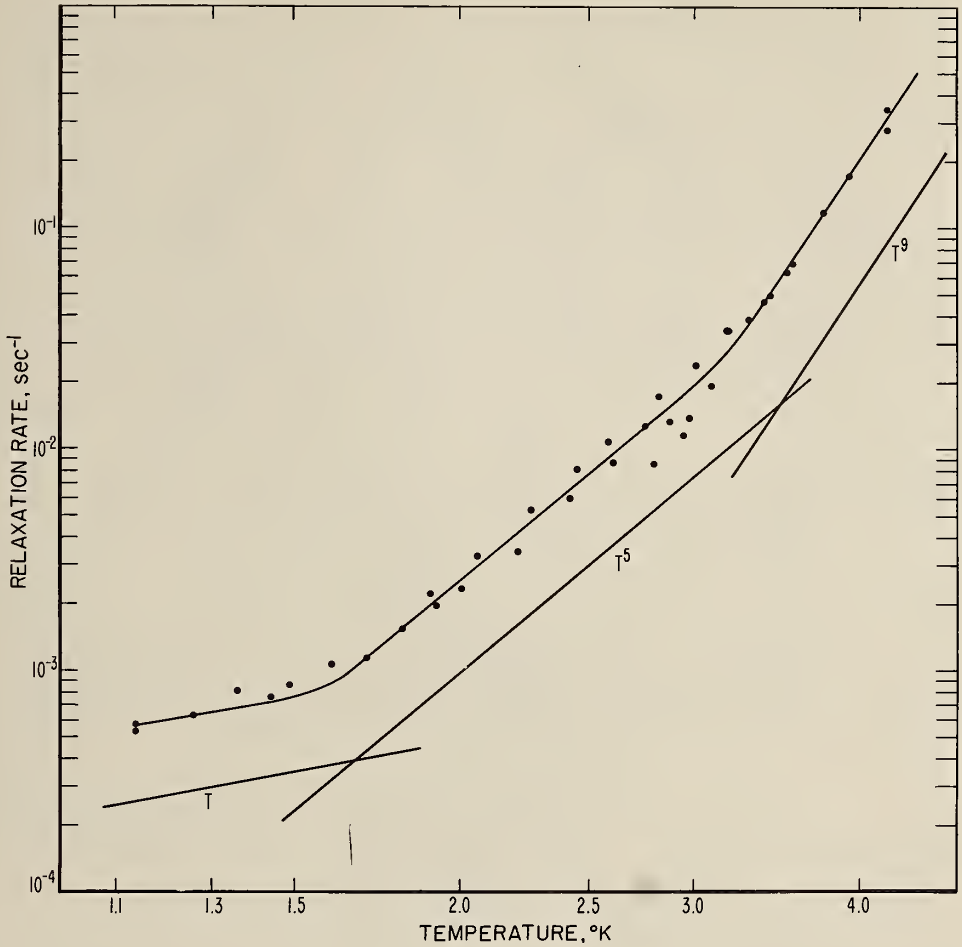

Figure 4 - The temperature dependence of the relaxation rate of $Mo^{5+}:TiO_2$. The dots are experimental points. The lines marked T, T^5, and T^9 are theoretical curves

4.6. The Measurement of Optical Frequencies and a Redetermination of the Velocity of Light by Microwave Modulation of Laser Beams (D)

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The method used is based on the principle, that if the difference and the ratio of two optical frequencies is measured, the frequencies themselves can be determined. A simultaneous measurement of the wavelengths then yields the velocity of light. The project consists of three phases.

I. Stabilization of the optical frequencies to be measured. The possibility of locking a He Ne 6328Å laser to the center frequency of the atomic transition by the use of loss modulation was experimentally demonstrated. The precision in the frequency lock was found to be better than 1 part in 10^{10}. In order to see and utilize the full possibilities of this new locking method, the building of two systems was started with the cooperation of the Electronic Apparatus Design Section. This developmental work is now nearing completion.

II. Modulation of the stabilized laser beam to produce sidebands whose difference frequency is known. For accuracy, the highest possible modulation frequency is required. The technique first applied by R. Targ, G.A. Massey and S.E. Harris[1] at 3 GHz was extended to 25 GHz.

An intracavity electro-optical loss modulation was applied to a 6328Å He Ne laser by the use of a KDP crystal excited at microwave frequencies of 10 GHz and 25 GHz.

The crystal was oriented with the c axis nearly parallel to the direction of the optical beam propagation, the a and b axes parallel and perpendicular to the beam polarization, respectively. The crystal was placed in a tuned microwave cavity with the electric vector along the c axis. In this position a part of the energy of the optical beam was converted to the opposite polarization and was coupled out by reflection at the Brewster window.

The sidebands, $\nu + \omega$ and $\nu - \omega$, (where ν is the optical and ω the microwave frequency) so generated were observed visually in the ring system of a Fabry-Perot interferometer. For the measurement of the intensities in the sidebands relative to that of the main band, a scanning interferometer was used with a low frequency ($\sim$ 50 Hz) sweep over the transparency curve. The intensity ratios were determined by using a photomultiplier the output of which is phase-detected against the sweep.

Preliminary experiments showed that the energy converted to the sidebands was a few tenths of a percent of the output of the laser for a one watt microwave excitation. It was shown that one can easily obtain 10^{11} photons per sec (10^{10} photoelectrons per sec with commercial photomultipliers) for two tenths of a watt microwave excitation with a laser of about 50 cm in length. These figures are not thought to be an upper limit; there are several ways of increasing the energy output in the sidebands (for example, by the use of a Glan-Thompson prism). There was no indication in the preliminary experiments that the electro-optical response of the KDP was less at 25 GHz than at 10 GHz.

Quantitative experiments are underway to relate the observed effects directly to the Pockels-coefficients.

III. Our theoretical studies have shown that provided the order numbers are known, the ratio of the two optical frequencies can be determined by tuning a passive cavity (Fabry-Perot interferometer) to both sidebands. Preparations for experimental utilizing this technique is now under way.

It should be noted that the accuracy in any determination of the velocity of light is limited by the accuracy of the present wavelength standards to about 1 part in 10^8. Our optical frequency determinations are based on frequency standards of much higher accuracy (about a part in 10^{12}). Thus the possibility exists that future wavelength standards can be based on optical frequency measurements and on a value of the velocity of light, specified so as to be compatible with present length standards.

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[1] Targ, R., Massey, G.A. and Harris, S.E. (Proc. IEEE 52, 1247 (1964).

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