RESEARCH IN QUANTUM ELECTRONICS

Research Grant DAHC04-67-C-0023
from the
Army Research Office (Durham)

FINAL REPORT
October 1, 1966 to June 30, 1968

Prepared by
C. O. Alley, Principal Investigator

SPONSORED BY
Advanced Research Projects Agency
Project DEFENDER
ARPA Order No. 675

UNIVERSITY OF MARYLAND
DEPARTMENT OF PHYSICS AND ASTRONOMY
COLLEGE PARK, MARYLAND

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I. Summary of Research

A. Introduction

The grant helped to continue research in a number of areas of quantum electronics which had been initiated under a special ARPA supplementary grant to the principal investigator under the interdisciplinary materials science program. These areas and the research accomplishments are discussed below under separate headings. A NASA grant (NGR 21-002-022) also provided partial support for some of the research during the period covered by the ARO(D) grant.

As a direct result of the ARO(D) support eight graduate students completed the research requirements for the Ph.D. degree and four graduate students completed the research requirements for the M.S. degree. There have been fourteen papers published so far on the work which this grant helped to support.

Although the design activity associated with the successful Apollo Laser Ranging Retro-Reflector experiment (see e.g. C. O. Alley, et al, "Laser Ranging Retro-Reflector: Continuing Measurements and Expected Results", Science 167, pp 458-460 (1970) ) was not explicitly supported by this grant, the research atmosphere which it contributed to at the University of Maryland was a major factor in the successful development of the lunar ranging techniques.

B. Correlation Effects in Laser Radiation

Careful measurements were made in the threshold region of oscillation, extending the accuracy and range of other measurements, as well as measuring higher order correlations. The use of cumulants,
which measure directly photon bunching with lower order correlations associated with accidental bunching removed, was introduced into the literature by this work. (See papers 5, 6, and 7 in section II and abstract of thesis of Ren-Fang Chang). Agreement with theory was established.

New theoretical techniques for the analysis of photon counting experiments were developed. (See papers 4 and 8 in Section II). These were used to analyze the possibility of investigating phase transitions by photon counting techniques. An experiment is in progress to examine the validity of the assumption of a single correlation length that is usually made in developing the dynamical scaling laws.

Another type of correlation concerns the spontaneous radiation from the upper or lower of two lasing states to a third state. A theoretical analysis of the line shapes to be expected was carried out, giving many details of the effect which has been confirmed by observations in other laboratories. (See abstract of the thesis of Romola Bose and paper 13). This effect serves to eliminate the usual Doppler width and allow the observation of optical transitions with only the natural line width. In combination with recent developments in the tuning of dye lasers and semiconductor junction lasers (See section G below) a new era of optical and infra-red spectroscopy is anticipated. The existence of the effect was recognized by J. A. White independently of other groups.
C. Scattering from Fluctuations in Optical Media

The technique of self-beating of light as pioneered by Forester and first applied to the study of phase transitions by Benedek was used to study the width of the Rayleigh line in laser light scattered from a solution of macromolecules (paper 1 in Section II) and from carbon dioxide at the critical point (See abstract of Thesis of John Osmundson and J. S. Osmundson and J. A. White, "Density Dependence of Inelastic Rayleigh Scattering in CO₂ near its Critical Point", Bulletin of the APS, II, 13, No. 2, p 183 (1968).)

These methods have been developed further and applied to the study of the critical mixing point in the binary fluid 3-methylpentane-nitroethane in collaborative work with Prof. Jan Sengers of the Institute for Molecular Physics of the University of Maryland. (See R. F. Chang, P. H. Keyes, J. V. Sengers, and C. O. Alley, Bulletin of the APS II, 16 No. 4, p 524 (1971).) The photon counting techniques discussed in section B above are being applied to the experimental study of this system. This work is now being supported by the Office of Naval Research.

D. Optical Radar Measurements of the Upper Atmosphere

Techniques were developed for detecting back-scattered light from a Q-switched ruby laser projected through a telescope pointing to the zenith. A rotating optical switch allowed the same telescope to serve as receiver. This was the first use of this method for an optical radar system. Enhanced back-scattering was observed at the height of the mesosphere (~ 80 km) at certain times. (See papers 2, 3, and 9 in Section II and abstract of thesis of Paul McCormick).
The proposed explanation of the sporadic enhancement of back scattering is the influx of debris from a comet when the earth passes through its plane (See abstract of thesis of Eric Silverberg). This explanation appears to be confirmed by the observation of enhancement by other optical radar groups at times predicted by our group. (See paper 15 in Section II).

As mentioned in the Introduction, the methods developed in this optical research were a major factor in the successful Apollo lunar ranging experiment.

E. Picosecond Laser Pulses

Considerable effort was devoted by Richard Smith to the analysis of precursor waves using the approach of H. Hoek in his Leiden thesis of 1938. This was the deduction of the extinction theorem by a careful examination of the local field including the near field of the oscillating molecular dipoles. This work has not yet led to publishable results. The search for a simple optical system in which to display and study precursor waves was not fruitful.

A new "linear" technique for measuring the autocorrelation of picosecond laser pulses was devised and demonstrated (See papers 10 and 11 of Section II and abstract of thesis of Richard Smith).

The techniques of working with picosecond pulses developed in the above work are now being used in an effort to extend the accuracy of the current lunar ranging from ± 1 nanosecond (± 15 cm) to ± 100 picosecond (± 1.5 cm).

An externally mode-locked He-Ne laser using acoustic modulation was constructed and studied. (See abstract of thesis of S. L. Chao).
F. Ring Lasers

Construction, operation, and study of square ring He-Ne lasers culminated in measurements on such a laser 20 meters in circumference, the largest yet constructed anywhere. Major emphasis was placed on mode competition and mode locking phenomena since the large dimension produces many modes within the line width and this leads to new effects. The conditions under which unidirectional behavior was exhibited were studied by changing isotopic mixtures and the magnetic field applied to the lasing medium. It was possible to reverse the direction of the propagating modes by reversing the direction of the longitudinal magnetic field. A theoretical analysis was made of the phenomena encountered. (See abstract of thesis of Richard Pohle).

Provision was made for flowing dry nitrogen around the ring and the shift of the beat frequency of two opposite propagating beams was studied as a function of the flow velocity. The possibility of measuring the second order Fresnel drag coefficient was analyzed but the requisite sensitivity was not easily obtainable.

The large ring laser readily exhibited a beat note of 365 Hz between oppositely propagating beams determined by the normal component of the earth's angular velocity. This is the optical analog of the Foucault pendulum. For the small rings constructed by our own and other groups, frequency entrainment between oppositely propagating modes occurs and no beat note due to earth rotation can be achieved.

G. Optical Pumping Studies of Rubidium and Cesium Vapors

A versatile experimental arrangement was developed for studying Zeeman and hyperfine resonances in the ground state of rubidium and
cesium atoms. The optical pumping light can be applied in controlled pulses as can the rotating Zeeman rf field and the microwave field at the hyperfine resonance frequency. (See abstract of thesis of Hai-men Lo and paper 12 in Section II). Spin echo type measurements of spin relaxation in buffer gas and small coated cells have been made. One interesting result was the verification that cells made in 1959 at Princeton by the principal investigator exhibited largely unimpaired relaxation times.

The pulsed triple resonance detection scheme for the o-o hyperfine resonance was applied to cesium for the first time after having been used successfully for rubidium. (See abstract of thesis of Gurbax Singh).

Successful matching of a gallium arsenide junction laser to the 8521 Å resonance line of cesium was achieved (See paper 14 in Section II and the abstract of the thesis of Gurbax Singh).

This is a development of considerable technical significance for the detailed study of light induced hyperfine frequency shifts and for the development of optically pumped cesium hyperfine atomic clocks and masers.

H. Stimulated Rotational Raman Scattering in Nitrogen

It has been planned to pursue research in this area. Initial exploratory experiments with a Korad K-2Q ruby laser were unpromising because of the large beam divergence of the laser. Therefore this research was not pursued further.
II. Published Papers


III. Scientific Personnel Supported and Degrees Awarded

A. Faculty

Partial salary support during summers and/or the academic year was provided to the following faculty members:

Robert Detenbeck, Associate Professor
Sherman Poultney, Assistant Professor
Philip Dilavore, Assistant Professor
Douglas Currie, Assistant Professor
Victor Korenman, Assistant Professor
Lester Erich, Visiting Professor (Lafayette College)

The research activities of C. O. Alley, Principal Investigator, and John A. White, Senior Research Associate, were supported by this grant although direct salary support was provided by other sources.

B. Graduate Students with Degrees Awarded and Thesis Titles

Romola Raychandhuri Bose, Ph.D., 1968
"Line Shape of Fluorescence from a Gas Laser"

Ren-Fang Chang, Ph.D., 1968
"Measurements of Temporal Correlations in the Electromagnetic Field Radiated by a Laser in the Threshold Region of Oscillation"

Paul D. McCoraick, Ph.D., 1968
"Optical Radar Measurements of Atmospheric Backscattering (40-100km)"

John S. Osmundson, Ph.D., 1968
"An Experimental Study of the Scattering of Laser Light from Carbon Dioxide Near the Liquid-Gas Transition Point"

Richard H. Pohle, Ph.D., 1969
"An Experimental Study of Large Helium-Neon Ring Lasers"

Eric C. Silverberg, Ph.D., 1970
"Interplanetary Dust Streams: Observation by Satellite and Lidar"

Gurbux Singh, Ph.D., 1971
"Optical Pumping Studies of Cesium-133: Phase Destruction Detection of Ground State Hyperfine Transitions and the Creation of Population Inversions with Gallium Arsenite Lasers"
Richard Smith, Ph.D., 1969
"A Linear Optical Technique for the Measurement of the Width of Ultrashort Optical Pulses"

Byong Hyup Ahn., M.S., 1967
"An Investigation of Rayleigh Scattering in Macromolecules"

Shui Lin Chao, M.S., 1967
"Generation of Short Optical Pulses by Mode Locking of He-Ne Laser"

Hai-men Lo, M.S., 1967
"Spin Relaxation of Optically Pumped Rubidium in a Homogeneous Magnetic Field"

John McClure, M.S., 1969
"On the Possibility of Probing the Hyperfine Structure of the 6^3P_3/2 Excited State of Cesium ^133 with a Gallium Arsenide Laser"
ABSTRACT

Title of Thesis: Line-Shape of the Fluorescence from a Gas Laser
Romola Raychaudhuri Bose, Doctor of Philosophy, 1968
Thesis directed by: C. O. Alley

The Weisskopf-Wigner theory of spontaneous emission from atoms
has been extended to include the possibility of stimulated transitions
through interaction of the atoms with an intense radiation field.
Expressions have been derived for the probability amplitudes of the various
states involved in a cascade of atomic excitations and decays, taking into
consideration the strong coupling of a single atom with an initially popu-
lated radiation field. The theory is further extended to include the
effects of initial atomic excitations. Two particular modes of excitation -
namely, excitation by spontaneous decay from higher excited states, and
radiative excitation from an external source, have been treated specifically.
The natural generalization to include the effects due to more than one
non-stationary atom has then been made by performing an average over the
atomic velocities.

This model has been used to investigate the frequency distribution of
the intensity of the fluorescent radiation from a gas laser. Fluorescence
from both the upper, as well as the lower laser level have been studied.
Detailed calculations have been carried out to incorporate the effects of
atomic velocities, frequency correlations in successive transitions, exci-
tation processes, etc. on the line shape of the fluorescence. The general
case for arbitrary strength of coupling between the atoms and the laser
field is worked out first and later the limit of weak coupling is also explored.

The calculated intensity profiles confirm experimental evidences of the modification of the shapes of spontaneous emission lines due to the influence of an intense radiation field. It is found that depending on whether the fluorescence originates from the upper or the lower level of the laser transition, a pair of symmetrically situated "holes" or "ears" (respectively) appear super-imposed on the Doppler broadened frequency distribution. The individual members of this doublet are found to be free of any Doppler broadening, their widths depending on the natural lifetimes of the lasing and fluorescing states only. However, an asymmetry exists between the widths and heights of the two different components of the structure. The shape, size, and positions of the super-imposed structure have been calculated in our theory for each of the cases mentioned above. There is excellent qualitative agreement of the theoretically derived expressions with the experimental results.
ABSTRACT

Title of Thesis: Measurements of Temporal Correlations in the Electromagnetic Field Radiated by a Laser in the Threshold Region of Oscillation

Ren-Fang Chang, Doctor of Philosophy, 1968
Thesis directed by: C. O. Alley, Associate Professor of Physics

In this study, we investigated the intensity fluctuations in a radiation from a laser in the threshold region of oscillation, which spans from 1/10 of threshold intensity to ten times threshold intensity. All experimental results are compared with theoretical predictions based on solutions of a Fokker-Planck equation.

We stabilized the intensity of a He-Ne cw gas laser against long-term intensity variations by means of a long time constant servo system which controls the cavity length, yielding a stability on the order of 0.1%. Precautions were taken in the isolation of the laser from external disturbances caused by building vibrations and acoustic coupling to mechanical resonances of the cavity.

Using a photomultiplier as detector, we analyzed the photocurrent with a spectrum analyzer and the photoelectron statistical distribution with a fast counting system. We measured the excess noise (in addition to shot noise) in the spectrum. We also calculated normalized cumulants of the intensity probability density function (IPDF) of the field from the photoelectron distribution measurement. The normalized cumulant is a true
measure of correlations among photons (known as photon bunching) because the contributions from lower order correlations (accidental bunching) are removed.

We found that the spectrum can be approximated by a single Lorentzian. The half-width varies from about 20 KHz at about one-tenth threshold intensity to a minimum of about 4 KHz at about twice threshold, and back to about 20 KHz at about 10 times threshold. The zero frequency component of the excess noise increases as the half-width decreases and vice versa. The agreement between the theory and the data is very good.

The statistics of the photoelectron distributions show that the intensity fluctuations at about one-tenth threshold are nearly those of a Gaussian field, and the IPDF approaches continuously the form appropriate to nearly constant amplitude when the intensity is brought up to about 10 times threshold. The transition in statistics from Gaussian at below threshold to constant amplitude above threshold is very well demonstrated by the data, which also agree remarkably well with the theory based on the Fokker-Planck equation.

The normalized 2nd, 3rd, and 4th cumulant of IPDF of the laser light were also measured at 17% and 42% of threshold intensity as counting time was increased from 3 microseconds to 1000 microseconds. The results agree with predictions computed under the assumption that the dependence of the correlation functions on the time variables is the same as for Gaussian light.
ABSTRACT

Title of Thesis: Optical Radar Measurements of Atmospheric Backscattering (40-100 km)

Paul D. McCormick, Doctor of Philosophy, 1968
Thesis directed by: C.O. Alley, Associate Professor of Physics

The design, development and operation of a laser optical radar system used for the detection of atmospheric backscattering from altitudes above 40 km is described in detail. A single optical path, for both transmission and reception of photons, is achieved by firing the laser pulse through an aperture in a rotating disc that serves as a "transmit-receive switch." The source of the photons is a pulsed ruby laser, operated in the Q-switched mode, with an energy output of 1-5 Joules per pulse and a pulse length of about $1.5 \times 10^{-8}$ seconds. Detection of backscattered radiation, at the single photoelectron level, is achieved by use of a 20 inch diameter Broken Cassegrain telescope (300 inch effective focal length), a photomultiplier tube and a pulse height discriminator. Data acquisition is done by using digital counting and logic circuits as an interface for an on-line digital computer which stores the accumulated pulse counts, in each altitude interval, for an arbitrary number of laser firings. Atmospheric backscattering profiles are obtained with an altitude resolution of 2.5 km.

Data obtained with the optical radar system is presented and analysed in detail. The returns from 40 km to about 65 km are in good
agreement with the theoretical molecular curve. Observations during the period January 23 - February 7, 1967 indicate a statistically significant enhancement of atmospheric backscattering (compared to the return predicted by the 1962 U.S. Standard Atmosphere) in the altitude range 77-82 km on the nights of February 4-7, 1967. The differential backscattering function, for this altitude range and on these nights, is found to be about $2 \pm 1 \times 10^{-12}$ cm$^{-1}$ ster$^{-1}$ which is approximately a factor of 20 above the molecular prediction.

It is concluded that the observed backscattering can be accounted for by an accumulation of aerosols (dust) near 80 km with a concentration and size distribution that is compatible with that observed by techniques other than optical radar. No backscattering enhancement was observed on the nights of January 30-31, 1967 from this altitude region.

Except for one statistically significant return (from 120-130 km) which was observed on February 7, 1967 in a profile based on 50 laser firings, no echoes were detected from altitudes above about 85 km. The system is not sensitive enough to detect molecular scattering at these altitudes.

A discussion of the noise sources inherent in an experiment using pulsed ruby lasers and sensitive photomultipliers is given.

The statistical interpretation of data involving the detection of small numbers of photoelectrons is stressed and discussed in detail.
ABSTRACT

Title of Thesis: An Experimental Study of the Scattering of Laser Light from Carbon Dioxide Near the Liquid-Gas Transition Point

John S. Osmundson, Doctor of Philosophy, 1968

Thesis directed by: C. O. Alley, Associate Professor of Physics

Light from a He-Ne gas laser was directed onto carbon dioxide held near the liquid/gas critical point. Light scattered at an angle of 11° with the forward direction was collected and analyzed by using a homodyne detection system.

Density of the carbon dioxide was measured to approximately ± 0.2% by measuring the height of the meniscus as a function of temperature when the phases had separated. Temperature of the CO₂ could be controlled to better than ±5 × 10⁻⁴°C for several hours. The optical cell containing the carbon dioxide was connected to a density control system which allowed measurements to be made at the critical density and over a range of densities from approximately \( \rho = (1 - 0.12)\rho_c \) to \( \rho = (1 + 0.09)\rho_c \) where \( \rho \) is the density and \( \rho_c \) is the critical density. Measurements were made at values of \( T - T_s \) (where \( T_s \) is the separation temperature for a given density ranging from 10⁻³°C to 1.0°C. The design and operation of the entire experimental apparatus is described in detail.

Data obtained with the experimental apparatus is presented and analyzed. For a critical density the linewidth of the power spectrum of the scattered light was proportional to \( (T - T_c)^{0.7} \), where \( T_c \) is the critical temperature; in addition the isothermal compressibility for
the critical density was proportional to \((T - T_c)^{-1.35}\) and the thermal conductivity then showed a \((T - T_c)^{-0.65}\) dependence. The linewidth also showed a density dependence of approximately \(|(\rho - \rho_c)/\rho_c|^{1.9}\). At large values of \(|(\rho - \rho_c)/\rho_c|\) the isothermal compressibility showed a much weaker dependence on \(T - T_c\).

The critical parameters \(\gamma\), \(\beta\), and \(\delta\) were measured. The best values were \(\gamma = 1.35 \pm 0.05\), \(\beta = 0.33\), and \(\delta = 4.9 \pm 0.2\). These values are in agreement with predictions of scaling law analysis.

(The critical constants \(\gamma\), \(\beta\), and \(\delta\) are defined in the usual way by the equations:

\[ K_T = \left(\frac{T - T_c}{T_c}\right)^{-\gamma}, \quad \rho = \rho_c, \quad T > T_c, \]

where \(K_T\) is the isothermal compressibility;

\[ (\rho_L - \rho_G) = \left(\frac{T_c - T}{T_c}\right)^{\beta}, \quad T < T_c, \]

where \(\rho_L\) and \(\rho_G\) are the densities of the liquid and gas phases respectively on the coexistence curve; and

\[ |P - P_c| = |\rho - \rho_c|^{\delta}, \quad T = T_c, \]

where \(P\) is the pressure and \(P_c\) is the critical pressure.)
ABSTRACT

Title of Thesis: An Experimental Study of Large Helium-Neon Ring Lasers
Richard H. Pohle, Doctor of Philosophy, 1969
Thesis directed by: C. O. Alley, Associate Professor of Physics

The output of a square, 20 meter perimeter, rf excited He-Ne ring laser was studied under various conditions of lasing gas pressure, cavity tuning, excitation, and magnetic field applied to the laser medium. The laser mode spectrum, (using a He-Ne scan laser as a heterodyne spectrometer) and the simultaneous intensity fluctuations (from DC up to nanosecond pulses) of both traveling wave beams (TWB) were studied.

The beat from the Earth's rotation between the oppositely directed TWB was observed and found to be about 365 Hz as expected at this latitude. The exact beat frequency could vary from 10 to 50 Hz around 365 Hz depending on the laser environment and parameters.

When a He³-Ne²⁰ gas 6.25:1 mixture was used as the lasing medium, unidirectionality was exhibited; 1) for low power single mode operation within 10 MHz of the gain center, 2) for self-locked operation in which the oscillating mode separation was approximately equal to \( \frac{3}{4} \) the pressure broadened relaxation constant of the gas, \( \gamma_{ab} \), and 3) for high power oscillation (>20W output). Unidirectionality was observed with no long term favored direction. The ratio of intensities of the two TWB was improved by aperturing and was at times >1000:1. A small amount of Ne²⁺ in the gas or the application of a magnetic field of >280 G to a small part of the medium removed the laser unidirectional
property.

The sum of the intensities of the two TWB for a given excitation was constant (except at high frequencies for self-locked pulses). Except in the three cases noted above, the He-Ne laser was not unidirectional and each TWB competed vigorously for emission energy.

Self-locked pulses of width = 5 ns and period equal to the inverse of the oscillating mode separation were observed, together with their mode structures. The most stable self-locked configuration observed contained 4 or 6 modes with amplitudes decreasing as their oscillation frequency differed from the Doppler gain center. Several less stable self-locked mode structures with complicated waveforms were also observed. An analysis of the mode phase relationships of the stable mode configurations is also discussed.

The laser traveling wave modes appear to obey Lamb's self-consistent equations for standing wave modes except for the absence of the standing wave "mirror" hole burned into the gain curve. Using Lamb's equations and a condition of constant energy output from the sum of the two oscillating and competing TWB, an equation is derived which describes the temporal behavior of the oppositely directed TWB both for strong coupling (b > B) and weak coupling (B > b). The saturation quenching of a single mode TWB by its rival required approximately 0.5 sec; but the quenching of a TWB by a self-locked or high power TWB required only about 30 m sec. It is suggested that transfer of energy through sidebands couples like directed modes so that they behave like a single mode. The observed difference in quenching speed is then related to a difference in the effective coupling constant (b - B), between the (slow) single mode or (fast) many mode situation.

The laser cavity and laser tube design is also discussed.
ABSTRACT

Title of Thesis: Interplanetary Dust Streams: Observation by Satellite and Lidar

Eric C. Silverberg, Doctor of Philosophy, 1970
Thesis Directed by: Sherman K. Poulteny, Assistant Professor

The interplanetary dust data from the orbiting satellites carrying "microphone type" detectors are studied in detail. It is shown that the dust showers seen by these satellites are related to low inclination, periodic comets. The orbital parameters of the dust leaving each of the indicated comets have been calculated. The dust which leaves the comet nuclei in elliptical orbits near their aphelia can build up in the plane of the comet and create flux densities which agree with the satellite results. Other observed characteristics of the dust showers are explained very well by such an interpretation. The size of the dust particle which participates in each event is calculated. These sizes along with the momentum sensitivities of the instruments indicate particle densities generally below .5g/cc. The characteristics of the low density, high velocity population of particles tends to lessen the apparent discrepancy between the microphone type satellite data and the data from the penetration and collection experiments.

Certain aspects regarding the atmospheric entry of interplanetary dust are studied. The optical scattering cross section produced in the earth's mesosphere is calculated for
several models. It was found that the van de Hulst distribution of particles will not create a significant population of atmospheric aerosols. Young cometary particles, on the other hand, will be important for either of two cases. Showers of small, low density particles (which do not ablate), or large, high velocity particles (which fragment), may produce a cumulative scattering in the mesosphere which exceeds the molecular contribution.

The lidar data on the mesosphere is reviewed along with some data of high altitude resolution recently taken by the author. It is shown that lidar methods can be applied to the study of many cometary dust showers. The present lidar data varies considerably among the various investigators regarding the appearance of dust, but may be reconcilable if the aerosol features are close to the resolution of the systems. It is shown that the present mesospheric lidar data which imply the existence of aerosols are consistent with cometary dust showers.
ABSTRACT

Title of Thesis: Optical Pumping Studies of Cesium-133: Phase Destruction Detection of Ground State Hyperfine Transitions and the Creation of Population Inversions with Gallium Arsenide Lasers

Gurbax Singh, Doctor of Philosophy, 1971

Thesis directed by: C. O. Alley, Associate Professor of Physics

The motivation of the present investigation was to carry out an experimental study of several different processes with a view toward creating population differences in the ground states of alkali atoms. (Cesium-133 was specifically studied but the techniques developed are equally applicable to other alkali atoms). Such studies may eventually lead to the development of cell-type Cesium atomic frequency standards and cesium masers. Both RF excited resonance lamps and GaAs diode lasers were used for the present work. This dissertation is accordingly divided into two major parts.

Part one reports investigations which were made for the development of the triple resonance coherent pulse technique for the creation of population differences among the ground state levels in cesium-133 and for the detection of the microwave induced hyperfine transitions by destroying the phase relationships between the various ground state levels produced by a radio frequency (Zeeman) pulse.

Using this method we have successfully detected the hyperfine resonances of the ground state of Cs-133. In addition, we have succeeded in making parallel wall-coated Cs-133 vapor cells with very long relaxation times for optically pumped alkali atoms. (Relaxation times over 250ms)
have been observed in 2.5" diameter cells.) Moreover, these cells have
been found to be very stable and have not shown any detectable deteriora-
tion even after a period of one & a half years.

A 'pulsed Cesium resonance lamp' which was essential for the
success of the coherent pulse technique, was also developed. It is well
known that the rf discharge in a conventional vapor lamp begins in an
unpredictable way some time after the application of the rf power. The
resonance lamp designed and operated by us showed very clean and repro-
ducible switching characteristics and the formerly present time lag did
not exist.

Part two describes the details of the achievement of population
inversions among the hyperfine levels in the ground state of Cs^{133} by
optically pumping these atoms with radiation from a GaAs diode laser.
The laser output was used to monitor the populations in the two ground
state hyperfine levels as well as to perform the hyperfine pumping.

By varying the injection current, a GaAs laser, operated CW at
about 77^oK, was used to scan the 8521 A line of Cs^{133}. The intensity
of the resonance scattering from cesium vapor served as an indicator of
the populations of the two levels involved. Experiments were performed
both with neon-filled and with paraffin-coated cells containing the
cesium vapor. The Doppler broadened and pressure broadened line width
of cesium in the neon-filled vapor cell at 100 Torr was found to be
1300 MHz for the D_2\textsuperscript{-} transition.

It was discovered that the diode laser could easily be tuned by
manually adjusting the injection current to match either of the hyperfine
components of the D$_2$ optical transition. The laser mode could be held on either component for up to a few seconds by manually controlling the current. This indicates that automatic locking should be possible.

Possible future applications, including a re-study of light shifts, the construction of a cesium maser, and the physics of optical pumping with coherent light are discussed.
ABSTRACT

Title of Thesis: A Linear Optical Technique for the Measurement of the Width of Ultrashort Optical Pulses

Richard Smith, Doctor of Philosophy, 1969

Thesis Directed by: C. O. Alley, Associate Professor of Physics

The most significant result which we have achieved in this report is that we have demonstrated that a linear optical technique can be used for the measurement of the width of ultrashort optical pulses which are produced by a mode-locked laser. In this technique a train of pulses is split into two trains in a Michelson interferometer. By observing the time interval over which the pulses interfered we were able to obtain a measure of the width of the ultrashort pulses produced by a mode-locked Nd laser. Our results indicated the typical pulse width for our laser to be about 3 picoseconds. By looking at the overlap of consecutive picosecond pulses we found that the total extent over which we could detect interference was at least 80 picoseconds and that the overlap region for consecutive ultrashort pulses was at least 13 picoseconds.

We also observed that the interference of consecutive pulses decreased greatly from the beginning of a laser shot to the end. This could imply a large difference in pulse height for consecutive pulses in the last half of a laser shot.

One of the advantages of the linear method is that in principle it can detect the presence of a low level background pulse upon which the high intensity portion may rest. No evidence was seen that indicated the presence of such a background pulse for our pulse trains.
ABSTRACT

Title of Thesis: An Investigation of Rayleigh Scattering on Macromolecules
Byong Hyup Ahn, Master of Science, 1967
Thesis directed by: Professor C. O. Alley

A beam of 6328 Å light from a He-Ne gas laser is scattered by a dilute solution of polystyrene in cyclohexane. The line-widths of the scattered beam caused by the concentration fluctuations of the binary fluid have been used to measure the molecular weights of the homologous series of the polystyrene. The samples ranged in molecular weight from 51,000 to 860,000.

The equation for the half-width of the spectrum is:

\[ \Gamma_i = \frac{A k T \sin^2 \frac{\theta}{2}}{V_i M_i} \]

where:
- \( A \): a constant for the homologous series
- \( k \): Boltzmann's constant
- \( s^2 \): \( 4 \sin^2 \frac{\theta}{2} \)
- \( \theta \): forward scattering angle
- \( T \): the absolute temperature
- \( V_i \): the volume concentration of the solute
- \( M_i \): the molecular weight of the solute

The experiment involves the comparison of the line widths of the spectra. At the same temperature and the same angle, the ratio of the line widths has the form of:

\[ \frac{\Gamma_i}{\Gamma_2} = \frac{V_2 M_2}{V_1 M_1} \]

The experimental values agree with the theoretical values with a fractional error ranging from 14% to 31%. 
Title of Thesis: Generation of Short Optical Pulses by Mode Locking of He-Ne Laser

Shui Lin Chao, Master of Science, 1967

Thesis directed by: Professor Sherman Poulton

A mode-locked laser can function as an optical pulse generator that produces a periodic train of spiked pulses of nanosecond width. The short pulses in the present study were produced by 1) forced-locking the optical modes in the laser cavity through the introduction of a time-varying loss into the cavity, or 2), self-locking them by lowering the Q-value of the cavity. A 6328 Å He-Ne laser ($L = 274$ cm) was used for this study. The time-varying loss was provided by an X-cut quartz transducer attached to a fused quartz block. The standing wave pattern, established in the fused quartz by applying a R.F. voltage to the transducer, formed a diffraction grating which attenuated the light beam by scattering a portion of it into higher orders of the diffraction pattern. The Q value of the resonant cavity was altered through lateral-tilting of the end mirror.

Optical pulses with widths as narrow as 3 ns were observed corresponding to 6 oscillating modes in the cavity. The repetition rate for the self-locked pulses was found to be $2(c/2\lambda)$ (i.e., twice the axial mode spacing) when the cavity Q was high and output power high. For low output power (i.e., low Q), the repetition rate was found to be $c/2\lambda$. For the forced-locked pulses, the repetition rate was found to be nearly constant when the loss frequency was close to $c/2\lambda$. On the other hand, the peak output power was found to be critically dependent on both the frequency and strength of
the loss modulator.

Mode amplitudes, measured with a scanning Fabry-Perot interferometer, were found to fluctuate severely and rapidly owing possibly to air turbulence in the uncovered cavity even in the forced-locked operation. Another possibility is that small, relative vibrations between the five mirrors (2 end mirrors and 1 reflecting mirror for laser, and 2 mirrors for interferometer) were responsible for the variations in mode amplitudes. A symmetrical spectrum of mode amplitudes was observed only when the light intensity was weak, and then only for short periods of time.
Title of Thesis: Spin Relaxation of Optically Pumped Rubidium in a Homogeneous Magnetic Field

Hai-men Lo, Master of Science, 1967

Thesis directed by: Associate Professor C. O. Alley

Spin relaxation for rubidium atoms oriented by optical pumping has been studied in the presence of neon buffer gas and in cells coated with long-chain paraffins or silicones (Dri-Film). Radio frequency pulse techniques were used in this experiment. Provision was made for the selection of either \( F = 1 \) or \( F = 2 \) Zeeman resonances in \( \text{Rb}_8 \) for which the \( g \) factor is \( +1/2 \) and \( -1/2 \), respectively. Due to the signals corresponding to the transition of \( F = 1 \) states being too small, the relaxation times were measured for \( F = 2 \) transitions only. Optical detection of spin echoes was achieved and used in the transverse relaxation time measurement. The accuracy on \( T_2 \) measurement is improved by cutting off the pumping light while the spin echoes are monitored. The results for measurement on longitudinal and transverse relaxation times \( T_1 \) and \( T_2 \) for the oriented rubidium \( 87 \) in 5 cm of neon as buffer gas are found to be 19 msec and 42 msec respectively. Measurement for a cell containing natural rubidium in a 2" cell coated with SC-77 gives \( T_1 = 46 \) msec and \( T_2 = 14 \) msec. The systematic error for the measurements on \( T_1 \) and \( T_2 \) are estimated to be 5% and 10% respectively. Previous measurements\(^1,2\) on these two cells gave \( T_2 = 50 \) msec. The large change in \( T_2 \) for the coated cell may indicate the deterioration of the coating.

The details of the experimental method, instrumentation, and results are given together with the theory, design, and construction of a mag-
netically shielded solenoid used in this work. The solenoid consists of a main solenoid winding and two correction coils. The whole system is enclosed in three concentric magnetic shields which serve to screen the inner region of the solenoid from external magnetic fields. Optical pumping measurements of the field homogeneity in the central region of the solenoid are reported. The shields reduce the disturbance due to changing external fields by two to three orders of magnitude. A feedback system was designed for further suppression of this disturbance. It reduced the noise more than one order of magnitude. The solenoid-shield system was at 0.143 gauss a homogeneity of 5 parts in $10^5$ over a $2''$ diameter sphere at the center of the solenoid.

With the addition of microwave circuitry the present optical pumping system can be easily used to study the ground state hyperfine transitions and the light shift effect. With some minor changes this can be converted into an optically pumped rubidium maser.