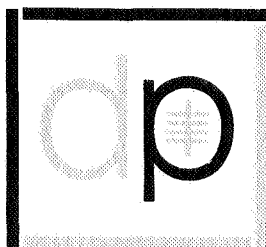


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COLUMBIA UNIVERSITY

DEPARTMENT OF PHYSICS

■ PROGRESS REPORT No. 14

MAY 1, 1966 THROUGH OCTOBER 31, 1966

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To:

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COLUMBIA RADIATION LABORATORY NEW YORK, NEW YORK 10027

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 ■ OCTOBER 31, 1966

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2 COLUMBIA RADIATION LABORATORY

3 RESEARCH INVESTIGATION DIRECTED TOWARD EXTENDING THE USEFUL RANGE OF THE ELECTROMAGNETIC SPECTRUM 4

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Prepared by R. Novick

1 COLUMBIA UNIVERSITY

Division of Government Aided Research

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9 October 31, 1966 10

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The names of the authors are arranged alphabetically.

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- B. Budick,² H. Bucka,³ R. J. Goshen,⁴ A. Landman,⁵ and R. Novick, "Fine and Hyperfine Structure of the 3^2P Term in Lithium," Phys. Rev. 147, 1 (1966).
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- H. M. Einbinder and S. R. Hartmann, "Oscillating-Field-Induced Magnetization in Solids," Phys. Rev. Letters 17, 518 (1966).

-
1. Present Address: General Telephone and Electronics Laboratories, Inc., Bayside, New York.
 2. Present Address: Microwave Laboratory, Physics Department, Hebrew University, Jerusalem, Israel.
 3. Present Address: Institut für Kernphysik an der Technischen Universität, Berlin, West Germany.
 4. Present Address: Hudson Laboratories, Columbia University, Dobbs Ferry, New York.
 5. Present Address: General Telephone and Electronics Laboratories, Bayside, New York.

Papers by CRL Staff Members Presented at Scientific Meetings

Second Rochester Conference on Coherence and Quantum Optics
(Sponsored by the AFOSR and the AFCRL), University of Rochester,
Rochester, New York, 22-24 June 1966.

S. R. Hartmann, "Photon Echoes."

International Conference in Solid State Science, American
University in Cairo, Cairo, U.A.R., 3-7 September 1966.

N. A. Kurnit and S. R. Hartmann, "Recent Photon-Echo Experiments."

Conference on Physics of Free Atoms, University of California,
Berkeley, California, 12-14 September 1966.

W. Happer, "Measurements of the Relaxation Times of the Magnetic
Dipole Moment and Electric Quadrupole Moment of Excited
Atomic States"; "A Computer Program to Evaluate the Breit
Formula for Level-Crossing Experiments."

R. C. Isler, S. Marcus, and R. Novick, "Fine and Hyperfine
Structure of the 3^2P State of Li^6 ."

G. Sprott and R. Novick, "Radio-Frequency Spectroscopy of
Metastable Autoionizing Alkali Atoms."

19th Annual Gaseous Electronics Conference, Atlanta, Georgia,
12-14 October 1966.

S. H. Dworesky, R. Novick, W. W. Smith, and N. Tolk, "Optical
Excitation of Helium Atoms by Low-Energy Helium Ions,"
Abstract G2, p. 24.

CRL Resonance Seminars

Meetings are held weekly at Columbia University, New York, N.Y.,
during the academic year and are open to all members of the
Physics Department. Guest speakers are invited to discuss work
in the general area of the research in the Columbia Radiation
Laboratory.

- H. H. Michels, United Aircraft Corporation, "Recent Developments in the Calculation of the Electronic Structure of Diatomic Molecules and Ions," June 8, 1966.
- H. Kleinpoppen, Physikalisches Institut der Universität Tübingen, "Polarization and Excitation Cross Sections of Hydrogen and Hydrogen-like Lines by Electron Impact," August 29, 1966.
- H. Bucka, Institut für Kernphysik, Technische Universität Berlin, "Resonance Fluorescence Investigations of Excited Atomic Levels," September 23, 1966.
- G. zu Putlitz, Erstes Physikalisches Institut der Universität Heidelberg, "New Results of Optical Radio-Frequency Experiments in Alkalis," September 30, 1966.
- W. T. Walter, TRG, Inc., "Efficient Pulsed Gas Discharge Lasers," October 7, 1966.
- R. N. Sternheimer, Brookhaven National Laboratory, "Shielding and Antishielding of Nuclear Quadrupole Moments," October 12, 1966.
- B. L. Donnally, Yale University, "Polarized Particles from Metastable Hydrogen Atoms," October 14, 1966.
- G. C. Tisone, Joint Institute for Laboratory Astrophysics, Boulder, Colorado, "Detachment of Electrons from H^- by Electron Impact," October 17, 1966.
- A. G. Redfield, IBM Watson Laboratory, "Magnetic Crystallography and Nuclear Spin Relaxation in Type II Superconductors," October 21, 1966.
- A. H. Firester, Princeton University, "Light Modulation at the Ground-State Hyperfine Frequency of Potassium," October 28, 1966.

Lectures

- L. R. Wilcox, "Optical Scattering in Pure Fluids," Seminar, Yale University, New Haven, Connecticut, May 10, 1966.

- L. R. Wilcox, "Coherent and Quasi-Coherent Scattering in Carbon Dioxide," Seminar, Gordon Research Conference, Meridian, New Hampshire, August 29, 1966; "Optical Scattering in Pure Fluids," Colloquium, Lehigh University, Bethlehem, Pennsylvania, October 27, 1966.
- G. F. Sprott, "Metastable Autoionizing Atoms," Physics Seminar, Yale University, New Haven, Connecticut, October 19, 1966.
- R. Novick, "The Helium Negative Ion and Related Metastable Autoionizing States," Colloquium, State University of New York, Buffalo, New York, October 27, 1966.
- L. Blau, " ^3He , α) Reactions on ^{16}O , ^{39}K , ^{40}Ca ," Nuclear Seminar, Columbia University, New York, New York, October 28, 1966.

ABSTRACT

A striking interference pattern was found with the redesigned laser spectrometer when a laser beam was passed through a fluid near its critical point. It has been shown that important information can be derived from a detailed study of this pattern. Isotherms above and below the critical temperature can be deduced, and the shape of the liquid-gas coexistence boundary can be determined. The method has been applied in a study of xenon isotherms on a pressure-density plot. The analysis indicates that no fundamental departure from basic theory remains down to 0.001°C above the critical point. Thermodynamic parameters in the immediate neighborhood of the critical point have been measured with unprecedented precision.

Optical excitation of helium in low-energy (below 500 eV) He^{+} -He collisions has been observed. Studies of the very low intensity radiation emitted indicates that triplet states are favored over singlet states and that D and P states may be more strongly populated than S states. Also, a new source of positive helium ions with increased beam intensity at low energies has been developed.

Induced magnetic dipole transitions between the Zeeman sublevels of the 2^3P_2 , $F = 7/2$ state of singly ionized lithium have been observed at 50 MHz. These and other Zeeman resonance experiments at low frequencies and low magnetic fields provide a basis for subsequent measurements of the fine-structure separations and for an eventual redetermination of the fine-structure constant α .

The Zeeman transitions in metastable autoionizing atomic states of potassium have been thoroughly studied; in particular, the excitation functions and power dependence have been determined. Some direct hyperfine transitions were observed giving a $^4\text{F}_{9/2}$, $F = 6$ to $F = 5$ splitting of 568.8 ± 0.1 MHz. The apparatus has recently been modified to increase the accuracy with which the magnetic field can be determined, and thereby to obtain an improvement of two orders of magnitude over the previously reported measurements.

Experiments have been carried out to investigate the properties of off-resonant light as a probe of optically pumped vapors. Magnetic resonance was detected in an optically pumped rubidium vapor using light from the Sr II spectrum. No depumping of the vapor or attenuation of the probing beam could be observed.

1.3 ATOMIC PHYSICS

N67-26617

A. METASTABLE ATOMS AND IONS

1. Properties of the Metastable State of Singly Ionized Helium

a. Direct Detection of the Two-Photon Decay Mode*
(J. Artura, R. Novick, N. Tolk)

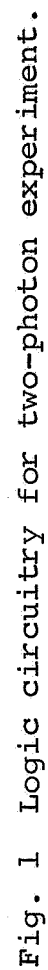
Coincident with a change in the personnel assigned to this project, an improved technique for studying the two-photon spectrum has been proposed. Instead of using two photon-detectors, three detectors will be installed: two "soft" uv phototubes (LiF windows) and one "hard" uv phototube (Be-Cu-O windowless).

A search will be made for two-photon coincidences between the "hard" uv tube and the "soft" uv tubes and also between the two "soft" uv tubes. The desired events should, of course, produce "hard-soft" coincidences but no "soft-soft" coincidences. The logic circuitry for performing these operations is illustrated in Fig. 1. High-speed pulse electronic equipment necessary for this experiment has been obtained. In order to improve the ion-beam intensity and hence the counting rate, a new ion source has been constructed⁽¹⁾ and will be installed during the next interval.

Program for the next interval: Using the three counting channels, we will continue work on the elimination of electronic crosstalk, which has plagued this work from the beginning. A search will then be made for two-photon coincidence signals in the spectral ranges of the "hard" and "soft" uv tubes.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) CRL Progress Report, October 31, 1966, p. 34.



b. Direct Lifetime Measurements*
(J. Artura, R. Novick, N. Tolk)

Work on the direct lifetime measurements has been further delayed in order to continue the two-photon experiment. A new ion source, developed in connection with the ion-atom experiment,⁽¹⁾ is being installed in the lifetime apparatus.

Program for the next interval: This program will be resumed upon completion of the two-photon experiment.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) CRL Progress Report, October 31, 1966, p. 34.

2. Metastable Autoionizing Atoms*
(L. M. Blau, M. Levitt, R. Novick, G. Sprott)

a. Rf Spectroscopy of Metastable Alkali Atoms

The observation of radio-frequency Zeeman transitions in metastable autoionizing states of potassium was described in the previous Progress Report.⁽¹⁾ Since then several characteristics of these lines have been investigated more thoroughly.

First, curves were obtained of the transition intensity versus the energy of the excitation electrons. The transition frequency and the strength of the magnetic field were held fixed on a resonant peak, while the output of the lock-in detector was monitored as a function of the electron-gun bombarding voltage. Simultaneously, the total metastable beam intensity was also monitored. Typical excitation curves are shown in Fig. 2. The similar shape of these two curves

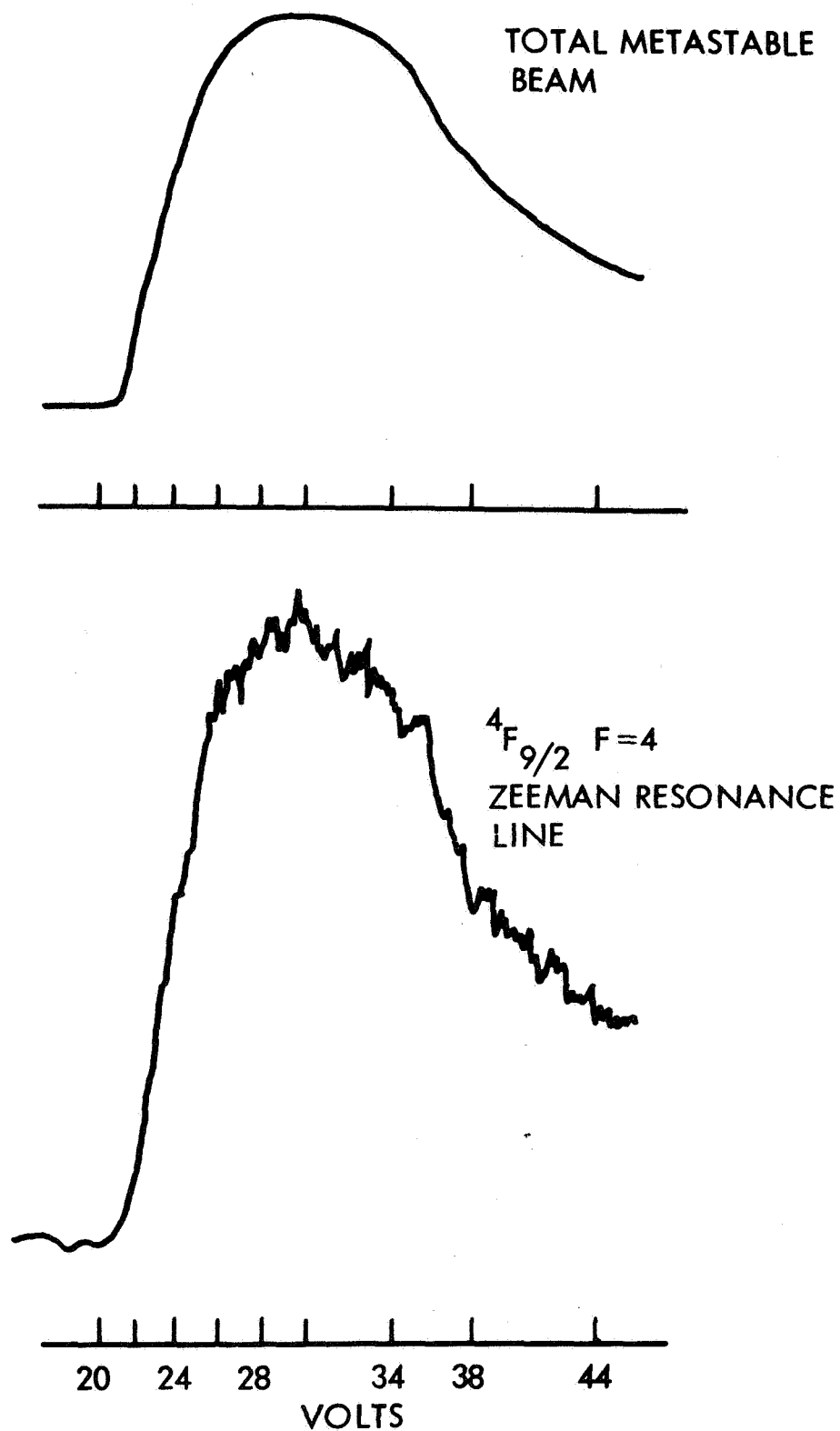


Fig. 2 Power dependence of line intensity
 $4D_{7/2}$, $F = 2$ Zeeman transitions.

verifies that the observed radio-frequency transitions definitely take place in the metastable state rather than in some other unexpected state of the atom. The measured excitation threshold is slightly higher than the previously determined metastable energy.⁽²⁾ A probable explanation is that the energy of the bombarding electrons is depressed by the presence of a large space charge in the excitation region of the electron gun.

Secondly, in order to investigate the high multiplicity of the observed lines, detailed power-dependence measurements were made of the four lines in the region where the $^4D_{7/2}$, $F = 2$, $m = -2 \rightarrow m = -1$ single Zeeman resonance was expected. Figure 3 shows a plot of line intensity versus rf power applied to the hairpin. The fact that each line appears at a power level about 10 times that of the previous line suggests that these lines are multiple quantum transitions⁽³⁾ (one to four quanta) from the $m = -2$ level to the $m = -1, 0, +1, +2$ levels. The positions and separations of these lines on a magnetic field sweep also support this explanation, but an accurate measurement of the transition field is needed for definite verification.

A search for direct hyperfine transitions revealed several which have been identified as transitions between the $^4F_{9/2}$, $F = 6$ and $F = 5$ levels. The extrapolated zero-field separation of these two levels is 568.8 ± 0.1 MHz. A search for the remaining lines was unsuccessful, probably because of insufficient rf power.

Unfortunately, until just recently the field in the C magnet could be determined only to the same accuracy as the

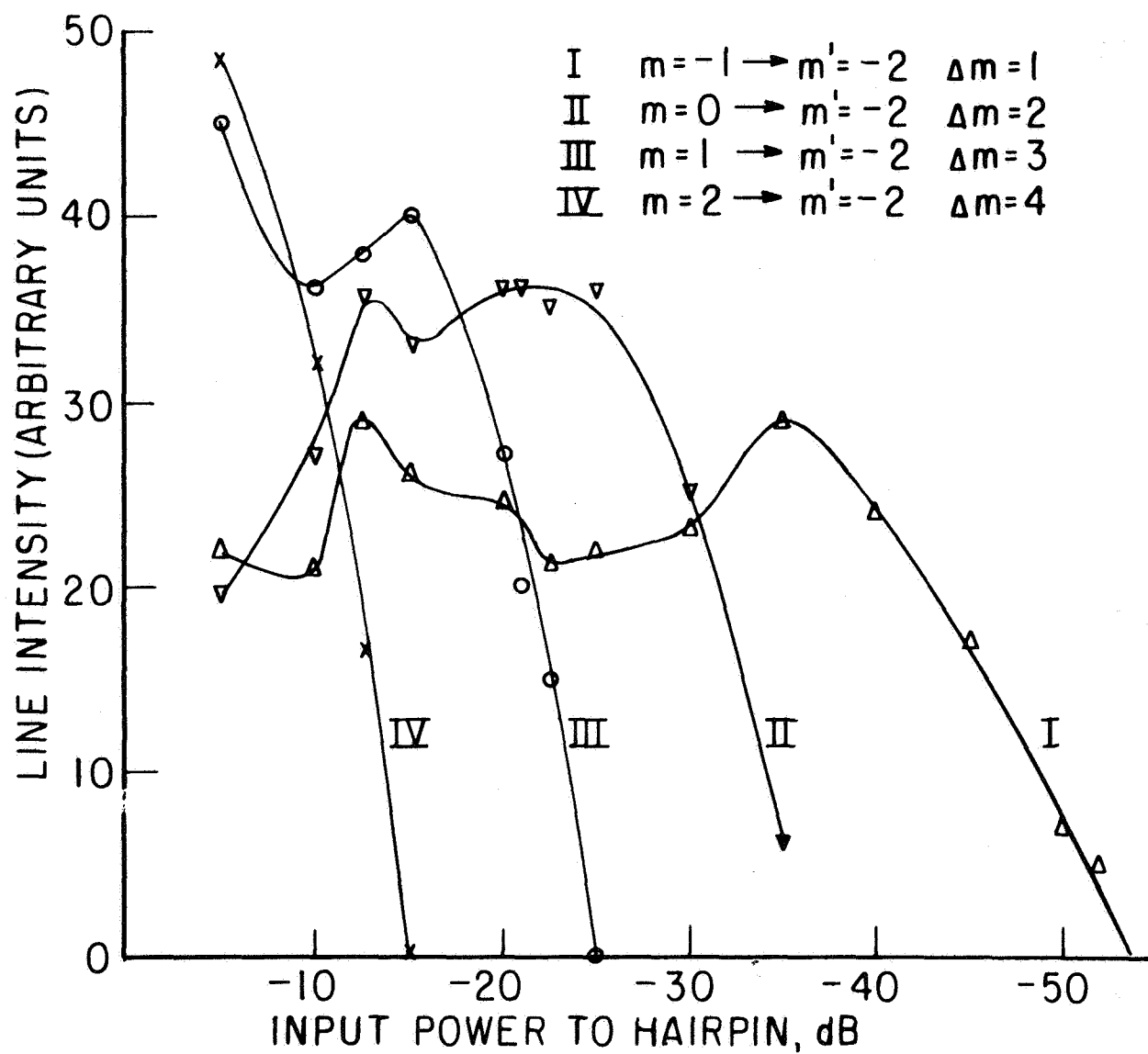


Fig. 3 Excitation curves of the $4F_{9/2}$, $F = 4$ resonance line and of the total meta-stable beam.

Hall probe used to monitor it, or about 2%. The usual accurate field calibration techniques employed in atomic beam research were unavailable because of the small dimensions of the apparatus and because the fields in the A and B magnets were not inhomogeneous. Therefore, in order to obtain an accurate calibration with Zeeman transition in the ground state of the potassium atom, the flat A and B magnet poletips were replaced by ones shaped to give inhomogeneous fields. With these new poletips, transitions can be seen in the potassium ground state by the standard deflection technique. Since the large magnetic fields in the A and B magnets still quench the metastable states, transitions can also be seen in the metastable state due to the differential quenching effect described previously.⁽⁴⁾

Modification of the apparatus has been completed, and Zeeman transitions have indeed been seen in both the ground and metastable states. The observed half-width of the ground-state transition is 240 kHz. This large half-width is due to the short transit time of an atom through the 1/8-in. hairpin. The center of the line can be determined to ± 5 kHz or better, which means that it should be possible to determine transition frequencies to less than five parts in ten thousand, an improvement of two orders of magnitude over our earlier measurements. With this increased accuracy, the previously observed transitions will be remeasured and a search will be made for additional direct hyperfine transitions. Once the potassium metastable state has been calibrated in this way, the other alkalis can be studied with the original homogeneous A and B field arrangement (which produces larger signals) using transitions in metastable potassium for calibration purposes.

b. Zeeman Quenching

The apparatus for measuring the Zeeman quenching of the metastable autoionizing $(1s2s2p)^4P_J$ states of lithium was successfully modified and is now being utilized. The principal components of the apparatus are a molybdenum oven, an electron gun, a movable detector, a Hall-effect probe, and an air-cooled proton NMR probe.

The molybdenum oven has a beam channel which is offset at an angle of 15° relative to an axis perpendicular to the direction of electron bombardment (which is parallel to the magnetic field). This geometry is utilized so that the metastable atoms, after suffering the recoil of the excitation impact, will travel along the mutual axis of the vacuum chamber and detector, while the other components of the beam will not pass through the collimating slits into the detector.

The electron gun consists of a barium-oxide-coated platinum cathode for copious electron emission (≈ 50 mA) and a control grid to regulate the electron beam passing through the metastable excitation region.

The detector can be moved a distance of about one metastable decay length (≈ 1 cm) along the chamber axis. Collimation at the entrance to the detector is provided by two Stark plates which serve to sweep out charged particles and to induce ionization of atoms in long-lived states lying just below the ionization limit. Electrons from metastables decaying in the detector are accelerated to a collecting plate by a small electric field. In order to reduce stray capacitance, which would cause attenuation of the modulated metastable signal, an electrometer triode tube is mounted in the detector near the collecting plate.

The Hall-effect probe and the air-cooled proton NMR probe are used to measure the magnetic field in the apparatus which is situated between the pole pieces of a 12-in. magnet. The NMR probe is used to measure fields below 4 kG and to calibrate the Hall-effect probe.

It was found that the gain of the detection system was independent of the magnetic field. Metastable signals observed by lock-in detection techniques were found to be several orders of magnitude larger than the noise generated in the electronic system of the detector. Several preliminary quenching curves have now been obtained with the existing apparatus. Figure 4 shows one of these quenching curves and a theoretical curve. The latter was obtained by calculating the various metastable decay rates as a function of magnetic field. The best available calculations of the level scheme and the matrix elements between the components of the discrete metastable states and the continuum were used. No explanation for the apparent discrepancy is presently available.

A dip in the curve was observed at a magnetic field of about 8 kG when a region several hundred gauss in width was swept at the rate of about 12.5 G/min with a lock-in time constant of 30 sec. A typical sweep is shown in Fig. 5. Data taken point by point in the region of this field are given in Fig. 6. The dip is not predicted by the present theory. However, it should be noted that the present theory does not take into account the effects of electron correlation, which are thought to be significant for the (1s2s2p) configuration.

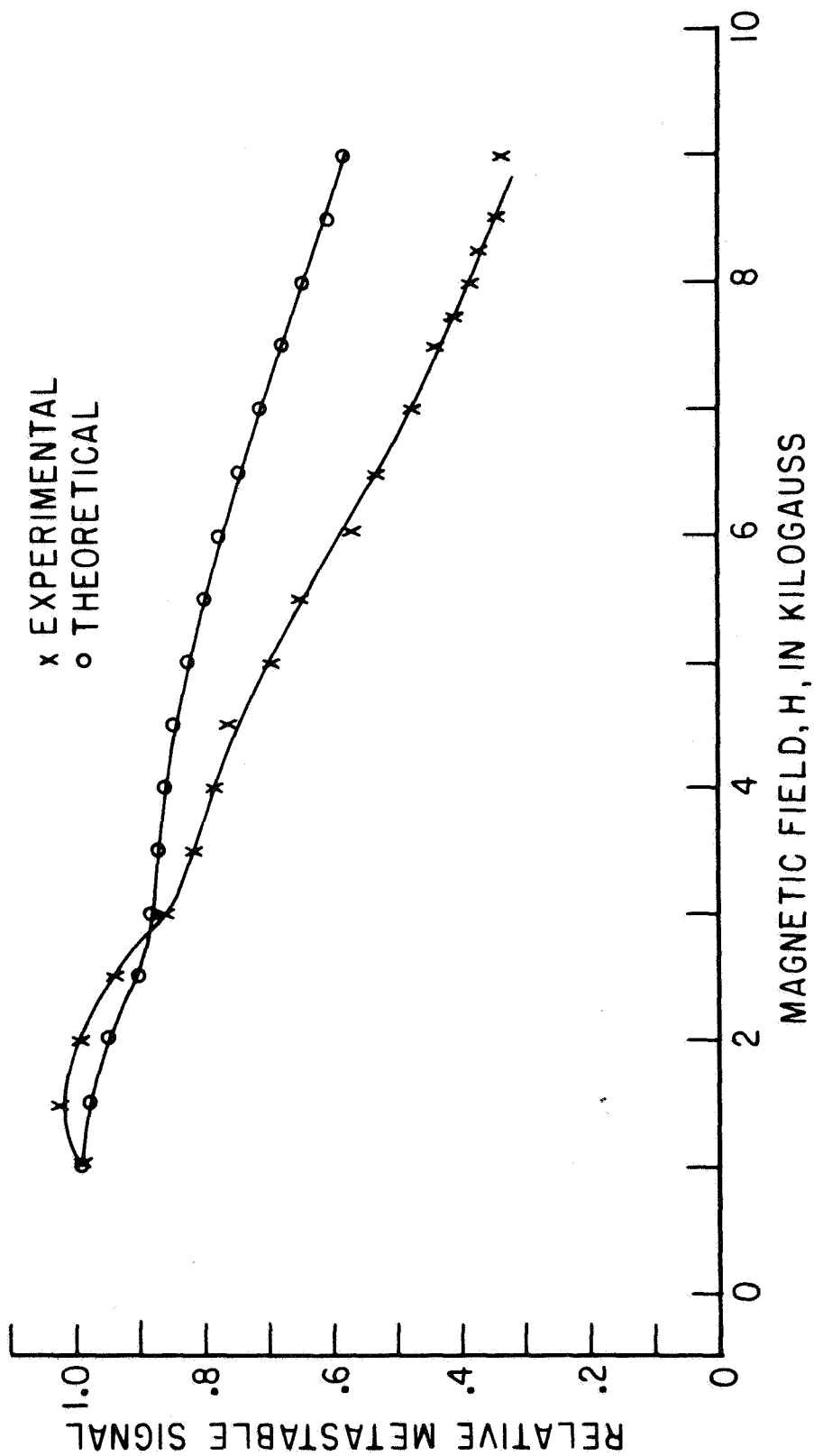


Fig. 4 Relative metastable signal as a function of magnetic field.

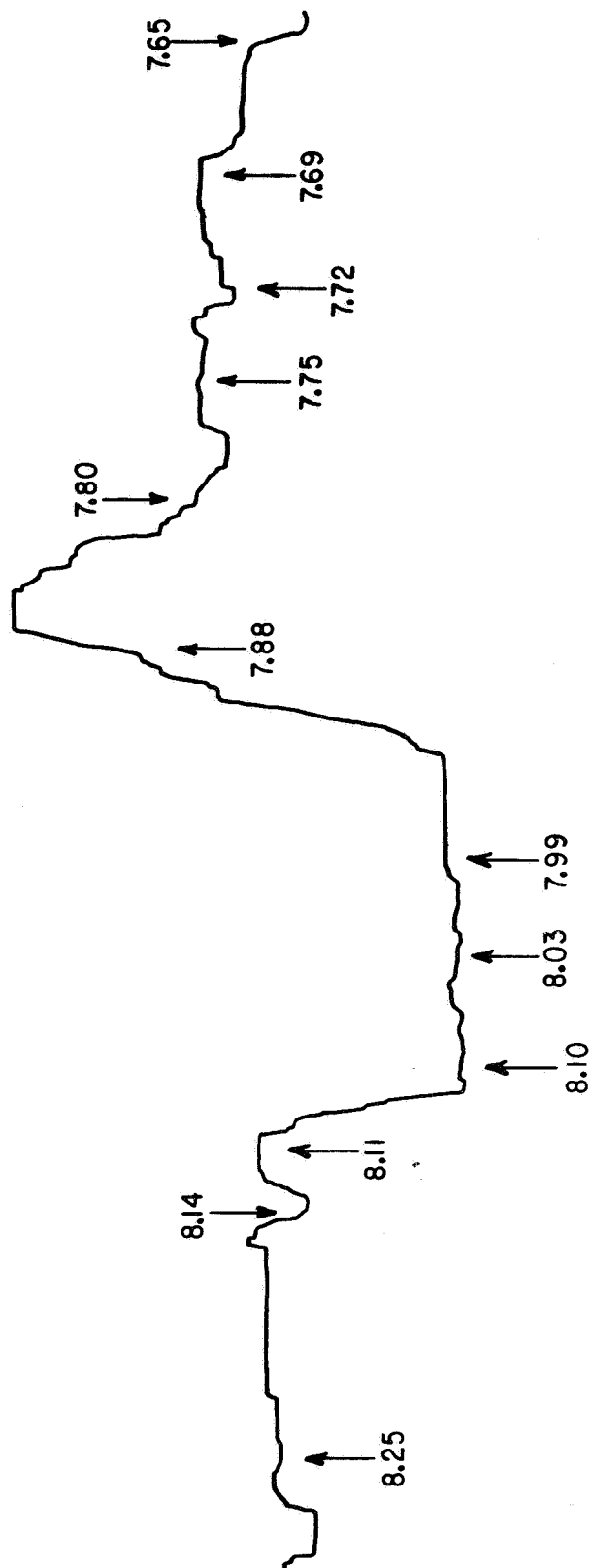


Fig. 5 Data on metastable signal obtained with magnetic field sweep.

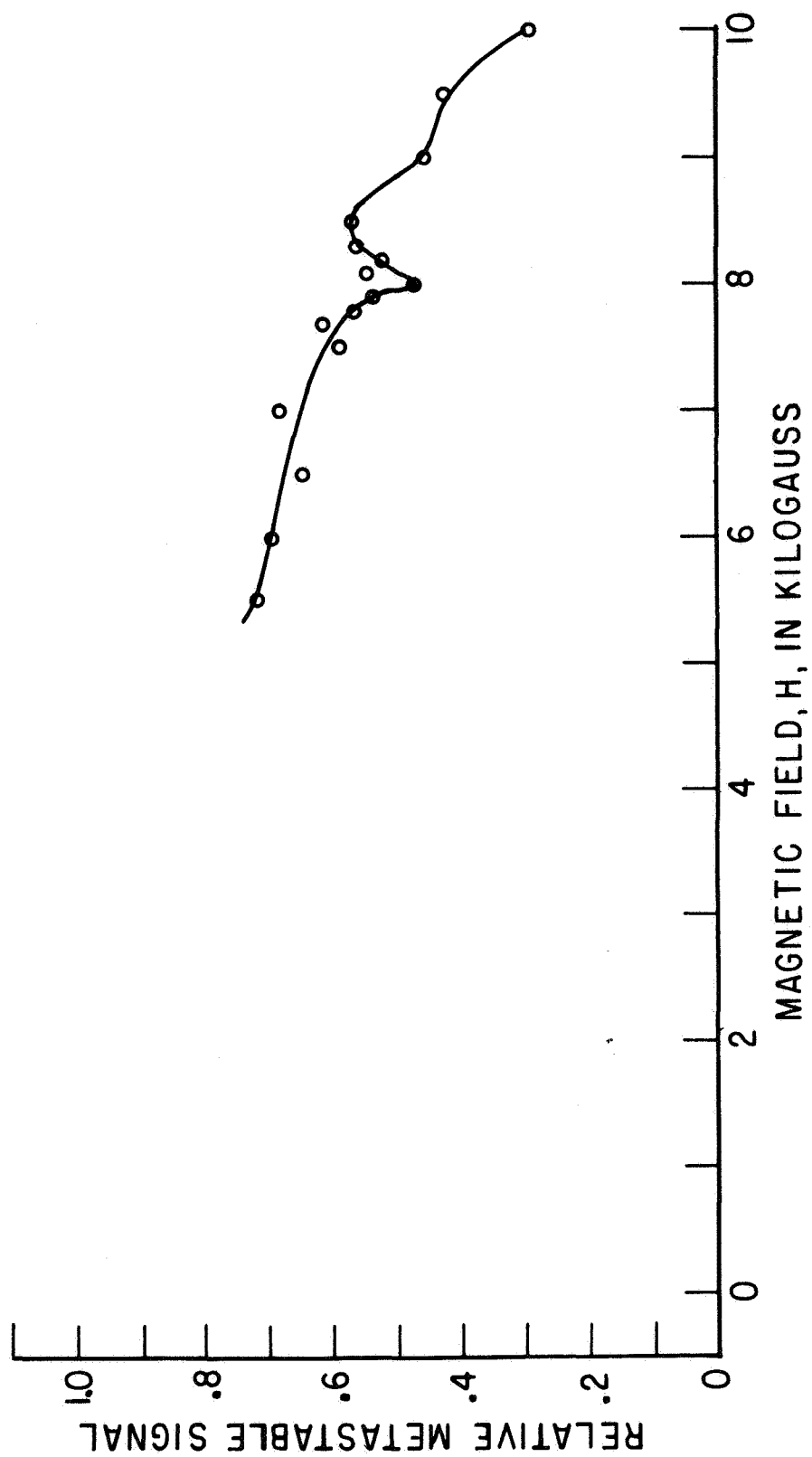


Fig. 6 Relative metastable signal as a function of magnetic field in region of 8 kG.

It is believed that the dip in signal at 8 kG is due to one of the two following phenomena:

1. Anticrossing (and hence, strong state mixing) occurs for two levels having the same magnetic quantum number but different admixtures of the eigenfunctions corresponding to the other principal quantum numbers, J and F. One level is composed predominantly of the longer-lived metastable eigenfunctions, while the other contains a greater percentage of the more rapidly ionizing components. The experimental observation that the dip is about 200-G wide allows one to predict from the theory for the anticrossing of two isolated levels that the distance of closest approach of the levels is in the range from $5 \times 10^{-4} \text{ cm}^{-1}$ to $10 \times 10^{-2} \text{ cm}^{-1}$ (15 to 3000 Mc/sec).

2. Transitions induced by a slight field inhomogeneity occur between two levels having different lifetimes and different magnetic quantum numbers. The field value for such transitions is near the point where the levels cross.

In either case, a low-frequency rf resonance experiment is suggested. If the effect in question involves the anticrossing of levels, there will be a minimum resonance frequency. By measuring the magnetic fields at which resonances are observed for various fixed frequencies, one can obtain the g values and identify the two levels. It is hoped that further studies will determine the lifetimes of the states and the fine and hyperfine structure splittings. An rf hairpin has been placed between the oven and the detector in the Zeeman quenching apparatus to enable immediate initiation of such studies.

c. He^- Ion

The theoretical investigation of the He^- system has been completed, and the results are as follows:

$$\begin{aligned} \text{(1) Lifetimes: } {}^4\text{P}_{5/2} &= 1,000 \text{ } \mu\text{sec} \text{ ,} \\ {}^4\text{P}_{1/2} &= 350 \text{ } \mu\text{sec} \text{ ,} \\ {}^4\text{P}_{3/2} &= 33 \text{ } \mu\text{sec} \text{ .} \end{aligned}$$

- (2) The calculations indicate that there is significant ($> 10\%$) mixing of the ${}^4\text{P}_{5/2}$ and ${}^4\text{P}_{3/2,1/2}$ states at low (≈ 1.0 kG) fields.

These new lifetime values indicate that our attention should be shifted from very short-lived components (≈ 100 nsec) to relatively long ones, while the results on mixing suggest that a careful investigation of Zeeman quenching in the region $H_z \leq 1.5$ kG should be made.

In order to measure these long lifetimes and to obtain magnetic fields ≤ 1.5 kG for investigating the Zeeman quenching of the beam, the 1-meter tube proposed previously⁽⁵⁾ has been replaced by a 10-meter tube which is presently being polished and will be wound with 1/4-in. copper wire. This will provide a low-field solenoid over the entire length of travel and allow us to measure the lifetimes of the beam components as a function of magnetic field.

The commercial rf ion source⁽⁵⁾ has been obtained from ORTEC (Oak Ridge Technical Enterprises Corporation) and far surpasses its original specifications as to current and to beam spread. Results of testing now being completed at CRL

indicate that at least 150 nA of He^- ion-beam current with an energy spread of 60 eV (full width at half maximum) at an energy of 5 keV can be obtained with careful tuning and aligning of the system. In order to observe the long lifetimes indicated by the theoretical calculations, a decelerating-transport-emersion lens system has been designed and is now being built. This lens will decelerate the He^- ion beam from 5 keV (the energy needed for the most efficient conversion of He^+ to He^- via collision with potassium vapor) to any energy between 10 eV and 1.0 keV. By combining this low velocity with the 10-meter length of the tube, we should be able to measure lifetimes in the millisecond region.

Almost all of the necessary supporting equipment has been assembled and the entire integrated system will be ready for testing and experimentation shortly.

Program for the next interval: The integrated system will be assembled and tested in its entirety, and actual experimentation on the He^- ion will begin.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

- (1) CRL Progress Report, April 30, 1966, p. 4.
- (2) CRL Progress Report, September 15, 1965, p. 15.
- (3) P. Kusch, Phys. Rev. 93, 1022 (1954); Harold Salwin, Phys. Rev. 101, 623 (1956).
- (4) CRL Progress Report, December 15, 1965, p. 4.
- (5) CRL Progress Report, April 30, 1966, p. 7.

TABLE I. Results of the present experiment and comparison with other experimental and theoretical work.

Nucleus	State	$2\beta^2 = E(\pm \frac{3}{2}) - E(\pm \frac{1}{2})$			Crossing Field (kV/cm) Present experiment
		A (Mc/sec)	B (Mc/sec)	$[\text{Mc/sec}/(\text{kV/cm})^2]$ Present experiment Other determination	
^{133}Cs	$7^2\text{P}_{3/2}$	16.609 ^a	-0.16 ^a	1.077±0.043 1.02, ^b theor.	11.07±0.2
^{85}Rb	$6^2\text{P}_{3/2}$	8.16 ^a	8.40 ^a	0.521±0.021 0.72, ^c exptl.	8.77±0.18, 11.0
^{87}Rb		27.63 ^a	4.06 ^a		14.0

^aG. zu Putlitz, Ergeb. Exakt. Naturw. 37, 105 (1965).

^bK. Murakawa and M. Yamamoto, J. Phys. Soc. Japan 20, 1057 (1965).

^cY. T. Yao, Z. Physik 77, 307 (1932).

B. LEVEL-CROSSING AND OPTICAL DOUBLE-RESONANCE SPECTROSCOPY

1. Electric-Field Hanle Effect

(W. Happer, A. Khadjavi, A. Lurio)

During this period we have measured the differential Stark shift of the $6^2P_{3/2}$ state of rubidium. Pure electric-field level crossings were used.⁽¹⁾ An energy-level diagram for the $6^2P_{3/2}$ state of Rb^{85} is shown in Fig. 7. In Fig. 8 the level-crossing signals observed in these experiments are compared with the theoretically predicted line shapes. The differential Stark shift (tensor polarizability) can be inferred from the electric field at which the crossings occur and from previously measured hyperfine interaction parameters. The results of our Stark-shift measurements obtained thus far for the alkali atoms are shown in Table I.

Program for the next interval: To measure differential Stark shifts and lifetimes of other excited states in the alkali atoms.

(1) A. Khadjavi, W. Happer, and A. Lurio, Phys. Rev. Letters 17, 463 (1966).

2. Fine Structure of Singly Ionized Lithium

(A. W. Adler, T. Lucatorto, R. Novick)

During this period rf-induced magnetic dipole transitions in the linear Zeeman region of the 2^3P_2 , $F = 7/2$ state of Li^+ have been observed. The 2^3P state of Li^+ was excited by electrons having an energy of about 30 eV above threshold directed along the Z axis. The measured polarization of radiation from the $2^3P \rightarrow 2^3S$ ($\lambda = 5485 \text{ \AA}$) transition, which provides an indication of population disparity among the

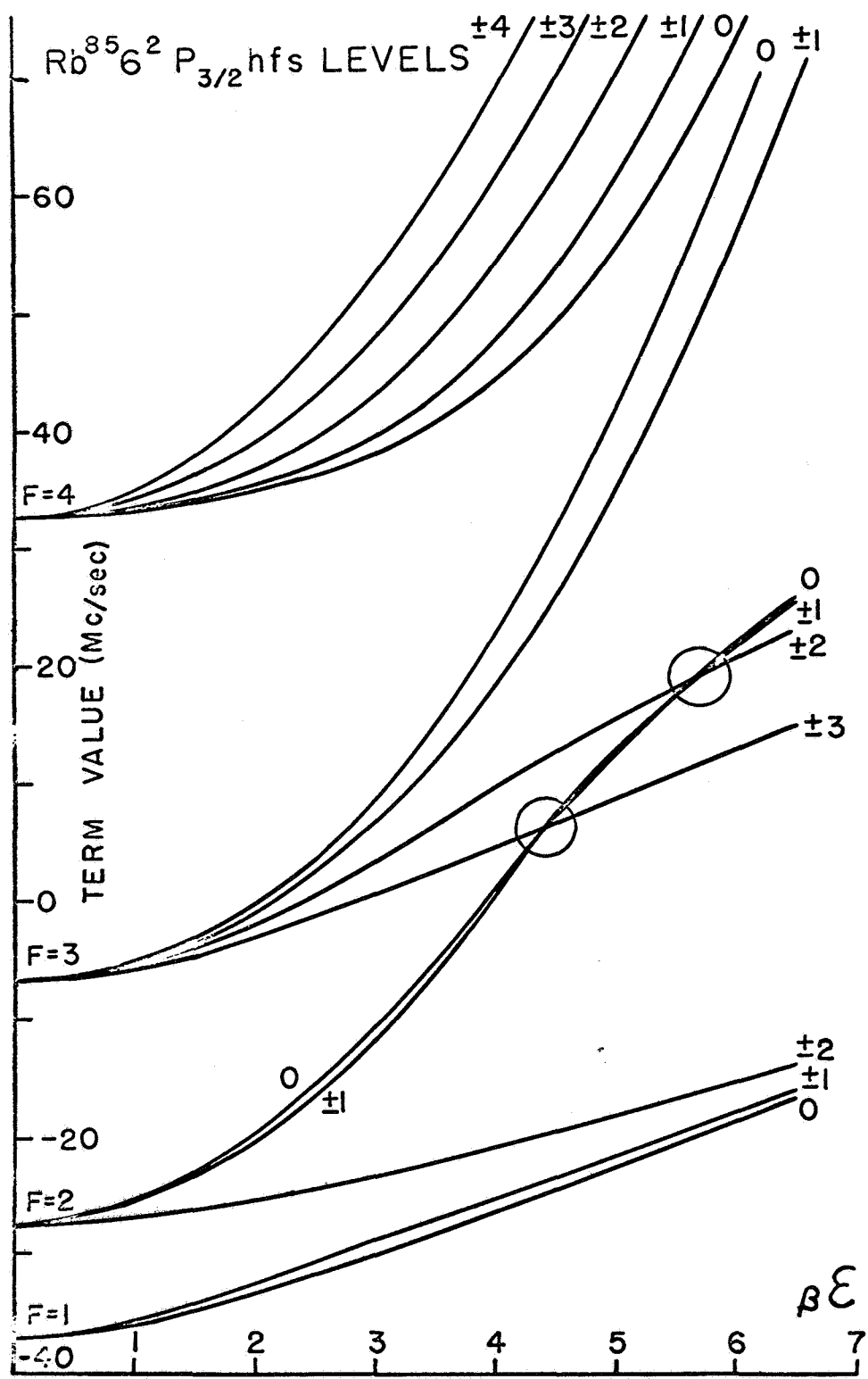


Fig. 7 Energy levels of $6^2P_{3/2}$ state of Rb^{85} in an electric field.

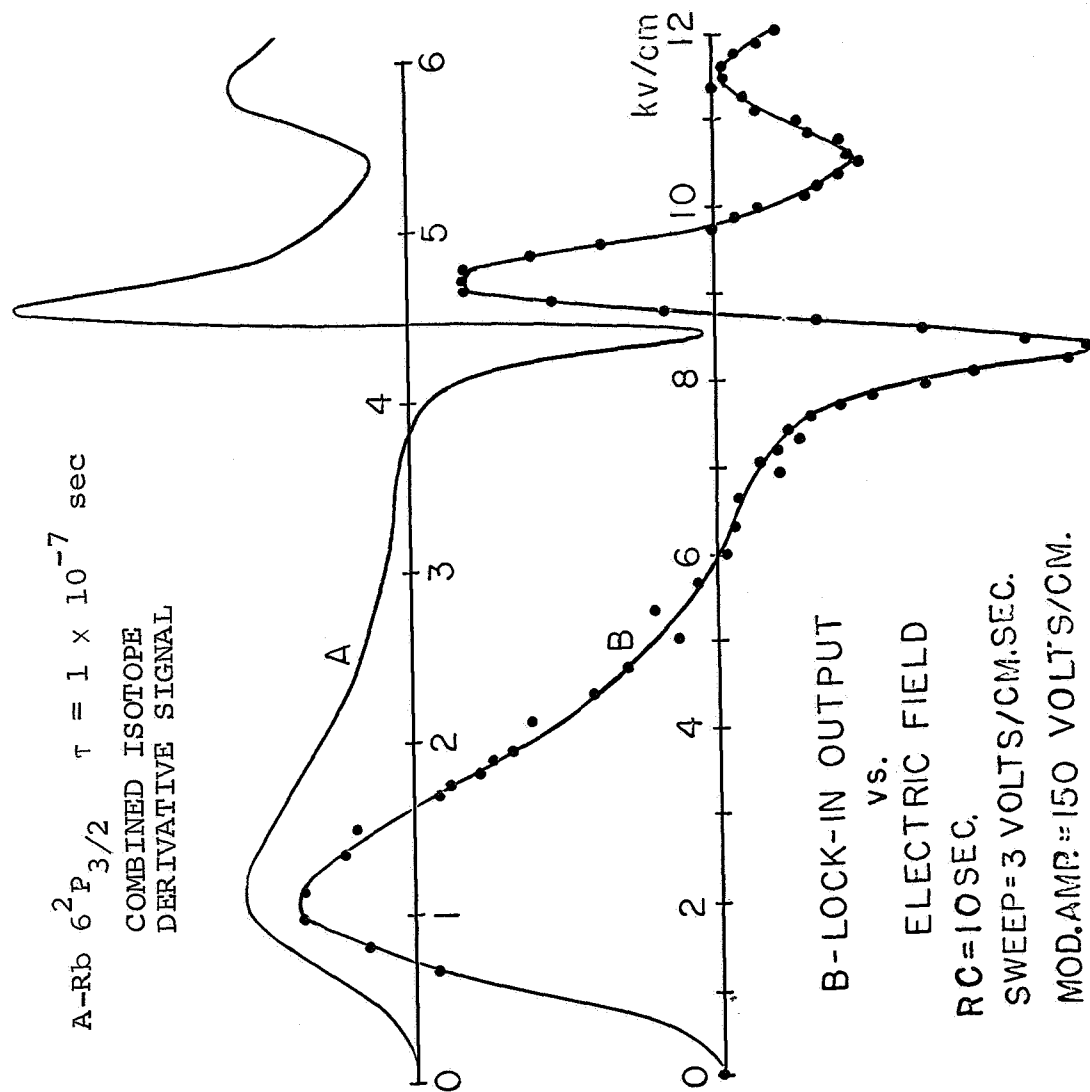


Fig. 8 Experimental and theoretical signals from Rb⁸⁵.

magnetic sublevels, was 2.5%. (The precise sublevel populations are readily calculable only in the case of threshold bombarding voltage, where the intensity of emitted radiation is too low to provide a measurable signal. The Oppenheimer-Penney theory gives a value of 10.7% at threshold.) An rf magnetic field of 1.2 G rms at 50 MHz was applied in the x direction. At resonance ($g_{7/2} = 1.2$ MHz/G, $H_{dc} = 41.7$ G), the rf field reduces the sublevel population differences of the $F = 7/2$ state, which results in a decrease in the polarization of emitted light. (In the threshold case an rf magnetic field of 1.2 G rms would cause a change in polarization from 10.7% to 10.4%.)

In order to detect a resonance against overlapping cyclotron resonance and other intensity fluctuations, a "double lock-in" technique was employed in which the rf power was rapidly turned on and off and a polarizer was slowly rotated about the line of sight. Digital integration techniques were employed and the preliminary observation exhibited a signal-to-noise ratio of about 3:1 after 5 hours of integration.

The ultimate goal of this experiment is to measure accurately the fine-structure energy separations by inducing magnetic dipole transitions between sublevels belonging to different J levels. Optical measurements by Herzberg and Moore⁽¹⁾ place these transition frequencies in the range between 60 - 160 GHz. A high-Q, spherical-mirror Fabry-Perot resonator has been constructed. Initial tests indicate that the magnetic field strength required for half-saturation can be obtained with about 2 W of input power.

Program for the next interval: 1) To optimize the experimental techniques for producing and observing induced magnetic dipole transitions at low frequencies, where the high power required is easily obtained. 2) To develop a high-Q cavity which will provide the electromagnetic fields required with the tubes of limited power which are available in the frequency range 60 - 160 GHz.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) G. Herzberg and H. Moore, Can. J. Phys. 37, 1293 (1959).

3. Hyperfine Structure of Group II Isotopes*

(A. Lurio, R. Novick, M. Swagel)

a. Barium $g_J(6^1P_1)$, $g_J(6^3P_1)$

The g_J values of the $(6s6p) \ ^1P_1$ and $\ ^3P_1$ states of barium were measured by optical double resonance in an atomic beam. The results are $g_J(\ ^3P_1) = 1.497 \pm 0.001$ and $g_J(\ ^1P_1) = 1.0039 \pm 0.0007$. The value for $g_J(\ ^1P_1)$ disagrees with a less precise measurement.⁽¹⁾ The sum of the experimental g_J values is 2.501 ± 0.001 as compared with the theoretical sum of 2.50116, including corrections for diamagnetic and relativistic effects. From the experimental g_J values, one can obtain the intermediate coupling coefficients c_1 and c_2 as shown in Table II.

With the $g_J(\ ^1P_1)$ value given in paragraph 1 and the value of the $(\frac{3}{2}, -\frac{3}{2}; \frac{1}{2}, \frac{1}{2})$ level-crossing magnetic field of Ba^{137} determined by Lurio,⁽²⁾ $A(\ ^1P_1) = 110.9 \pm 1.0$ MHz, which is in excellent agreement with the value $A(\ ^1P_1) = 108.2 \pm 3.0$ MHz obtained by Jackson and Tuan.⁽³⁾

Table II. Intermediate Coupling Coefficients.

<u>Method of Determination</u>	<u>c₁</u>	<u>c₂</u>
Lifetime of $^1P_1$ and $^3P_1$ States	0.5000	0.8660
Fine Structure	0.4938	0.8695
$g_J(^3P_1)$, $g_J(^1P_1)$	0.5040	0.8638
Pure L-S Coupling	0.5774	0.8165

It was found that the cell loading system was too unreliable to handle an expensive sample of Ba^{133} ; consequently, the experiment has been discontinued.

In any future experiments on $Ba^{133}CO_3$, the following improvements should be made: 1) The $Ba^{133}CO_3$ should be reduced inside the cell. This procedure would eliminate transfer loss; 2) The cell assembly should be designed to allow for the loading and bakeout time (no more than a day). This is compared with the present loading and bakeout time of two weeks. No further work on barium is contemplated at this time.

b. Mercury $g_J(^1P_1)$

An experiment is now under way to measure $g_J(^1P_1)$ for mercury by the level-crossing technique. Hg^{199} has $I = 1/2$; at approximately 2500 G, there is a $(\frac{3}{2}, \frac{3}{2}; \frac{1}{2}, -\frac{1}{2})$ $\Delta m = 2$ level crossing. The $g_J(^1P_1)$ and the crossing field are related by the equation

$$g_J = \frac{-A}{\mu_O H_c} - \frac{g_I}{2} ,$$

where A is the magnetic dipole interaction constant, and H_C is the crossing field. The A value has been measured precisely; the value of H_C will be determined in this experiment. The light path is flushed with N_2 to prevent absorption of the 1849-Å line by oxygen. The cathode of the Bendix electron multiplier is brass, which is much more photosensitive to 1849 Å than to 2537 Å. The lamp is cooled to maximize the 1849-Å output; the cell is cooled to -30°C to enhance the fluorescence at 1849 Å. The cell contains natural mercury which is composed of 16.9% Hg^{199} , 13.2% Hg^{201} , and the even isotopes of Hg. Fortunately, however, no level crossing in Hg^{201} occurs near 2500 G.

Fluorescence has been detected from the cell. The next step is to detect the Hanle effect and the level crossing. The $g_J(^3P_1)$ is already precisely known. From the values of $g_J(^1P_1)$ and $g_J(^3P_1)$, the values of the missing coefficients c_1 and c_2 can be calculated. Also the sum of $g_J(^1P_1)$ and $g_J(^3P_1)$ can be compared with $[g_J(^3P_1) + g_J(^1P_1)]$, assuming Russell-Saunders coupling, as a check.

Program for the next interval: A search will be made to detect the Hanle effect and the level crossing.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) H. Bucka and H. J. Schüssler, Ann der Physik 7, 225 (1961). The rf source used in this experiment was only 60 MHz; the Block-Siegart effect would not be negligible here.

(2) A. Lurio, Phys. Rev. 136, A376 (1964).

(3) D. A. Jackson and D. H. Tuan, Phys. Rev. Letters 11, 209 (1963).

4. Fine and Hyperfine Structure of the 3P State of Li^{7*} (R. C. Isler, S. Marcus, R. Novick)

During this period, data from the very low-field crossing experiment, described in the previous report,⁽¹⁾ were evaluated. This was done by comparing the observed line shape with that predicted with the computer program for various values of the hyperfine constants. Due to the poor signal-to-noise ratio, it was not possible to extract a b-value from the data. A value for $a_{\text{eff}}(3/2)$ for Li^7 was obtained, however, which is

$$a_{\text{eff}}(3/2) = -0.965(20) \text{ MHz} .$$

Using this value and the measured values of the hyperfine splittings in the higher field $\Delta m_J = 2$ crossing which were previously reported, we obtained for Li^7

$$a_{3/2 \text{ dipole}} = 2.11(3) \text{ MHz} ,$$

and

$$a_c = -3.08(3) \text{ MHz} .$$

The crossing field values reported previously⁽¹⁾ were also used to determine the central crossing field, and thus the fine structure separation, ΔW , which is

$$\Delta W = 2882.704(30) \text{ MHz} .$$

This value includes (a) corrections for departure from Russell-Saunders coupling, (b) the specific and normal mass-effect corrections to g_ℓ , (c) relativity corrections, (d) diamagnetic interactions within the atom due to the applied magnetic field, and (e) electrodynamic corrections to g_s .

Second differences in the crossing fields were calculated in an effort to evaluate the quadrupole moment of the Li^7 nucleus. Unfortunately, the second-order magnetic dipole correction which must be applied to these differences is very nearly equal to the electric quadrupole contribution and is opposite in sign. Thus any small error in $a_{3/2}$ dipole may lead to an enormous error in Q . Now the second-order corrections are based on at least one theoretical estimate which may be subject to change and which might produce a substantial modification of b . When writing the magnetic hyperfine Hamiltonian for a single electron in the usual way as ⁽²⁾

$$\mathcal{H} = -2\mu_o \left[-\frac{8\pi}{3}(\vec{s} \cdot \vec{\mu})\delta(\vec{r}) + \frac{(\vec{S} - \vec{L})}{r^3} \mu - \frac{3(\vec{s} \cdot \vec{r})(\vec{\mu} \cdot \vec{r})}{r^5} \right],$$

where $\vec{\mu}$ is the nuclear magnetic moment, it is tacitly assumed that $\langle 1/r^3 \rangle$ for the orbital part of the interaction is identical to $\langle 1/r^3 \rangle$ for the spin part. Recently, Harvey⁽³⁾ has reported experimental results which cast doubt on this assumption. Using an electron spin resonance technique on the ground and first excited states of O^{17} , he has measured four independent hyperfine constants which he was able to fit to three unknown parameters within the experimental error. These were $\langle 1/r^3 \rangle_{\text{orbital}}$, $\langle 1/r^3 \rangle_{\text{spin}}$, and core polarization effective $\langle 1/r^3 \rangle$.

In our analysis, we have assumed that $\langle 1/r^3 \rangle_{\text{orbital}}$ equals $\langle 1/r^3 \rangle_{\text{spin}}$, leaving two unknowns, $a_{3/2}$ and a_c . We were able to solve for these since we had two independent hyperfine measurements. If we consider $\langle 1/r^3 \rangle_{\text{orbital}}$ not equal to $\langle 1/r^3 \rangle_{\text{spin}}$, another measurement is required to determine the three unknowns. It can be seen by following

the analysis of Bethe and Salpeter⁽²⁾ that for diagonal elements, this modified Hamiltonian can still be written as $a_{jd}\vec{I}\cdot\vec{J}$. Direct evaluation of these elements indicates, however, that the relationship $a_{1/2d} = 5a_{3/2d}$ is no longer valid. We may thus restate the magnetic hyperfine problem in terms of the three parameters $a_{3/2d}$, $a_{1/2d}$, and a_c .

In an effort to obtain a third measured value, we have initiated a double-resonance experiment in the $3^2P_{1/2}$ state of Li^7 . This experiment should give us directly a value for $a_{\text{eff}}(1/2)$, which will enable us to solve for all of the hyperfine constants and detect any difference in the values of $\langle 1/r^3 \rangle_{\text{orbital}}$ and $\langle 1/r^3 \rangle_{\text{spin}}$.

It is clear that due to the present uncertainties in the corrections, we cannot obtain a reliable value for b and the related quadrupole moment Q . It is expected, however, that the results obtained from the double-resonance experiment will clarify the situation, and will yield a good value for b and Q . Until such time it is felt that we cannot reliably report these values.

A theoretical analysis was performed to estimate the effect of any nonlinearity in the spectral profile of the lamp over the crossing region. Assuming a Doppler profile over the crossing region, we calculated the effect to be negligible.

Experimental work was begun in a thus far unsuccessful effort to observe the double-resonance signal in the $3^2P_{1/2}$ state of Li^7 . The apparatus was the same as that used in the level-crossing work except that (1) radio-frequency power was introduced by means of an rf loop, (2) the incoming and

scattered light was circularly polarized, and (3) Helmholtz coils instead of the iron-core magnet were used to provide the constant magnetic field. This change was made in order to allow a component of the incoming light to be parallel to the magnetic field. This condition is necessary since, for optical excitation between two states of $J = 1/2$, it is impossible to excite the atom preferentially to one of the m_J levels by means of π light or undifferentiated σ light.

Program for the next interval: Further attempts will be made to observe the $3^2P_{1/2}$ double-resonance signal. In so doing, the following problems will have to be overcome:

(1) The circular polarizers which were used attenuated approximately 85% of the incoming 3233-Å resonance radiation. Better polarizers will have to be made or purchased. (2) The lenses of the apparatus are being clouded with lithium at an extremely fast rate, about twice as fast as when the apparatus was used for the level-crossing work. It is suspected that this is due to the presence of the rf loop, which is situated so as to deflect lithium atoms from the beam onto the lenses. This effect might be controlled by keeping the loop at a low temperature so that it will condense the lithium beam rather than deflect it.

Attempts will also be made to resolve the question of the discrepancies between our measured value of $Q(\text{Li}^7)$ and other measured values.

*This work was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) CRL Progress Report, April 30, 1966, p. 18.

(2) H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One and Two Electron Atoms (Academic Press, Inc., New York, N. Y., 1957).

(3) J. S. M. Harvey, Proc. Roy. Soc. (London) A285, 581 (1965).

5. Narrow-Linewidth Resonance Fluorescence (A. Lurio, R. Schmieder)

The interaction between an atom and electromagnetic radiation near the frequency of an atomic transition depends qualitatively upon the spectral character of the radiation. If the incident radiation has a linewidth that is broad compared to the natural linewidth of the atomic transition, the interaction is by single-photon absorption and atomic excitation, followed by atomic de-excitation and single-photon emission within the natural linewidth of the transition. If the incident radiation is nearly monochromatic, the energy of the intermediate state is practically indeterminate (from the uncertainty principle), and the system must be treated as a single, tightly coupled atom-radiation system. In this case, the incident and emitted radiation have the same line shape and are coherent; the emitted intensity is governed by the natural line shape.⁽¹⁾ Thus, a narrower-than-natural linewidth source provides a method for studying the natural line shape and the resonance interaction. Furthermore, it has the potential for forming a third test of quantum electrodynamics (after the anomalous electron magnetic moment and the Lamb shift). This is because the radiative corrections to the bound states⁽²⁾ result in a distortion of the natural line

shape of the order of $\alpha^2 \approx 0.6 \times 10^{-4}$. Measurement of this shift would provide an important confirmation of the theory.

An experiment has been initiated to investigate narrow-linewidth resonance fluorescence. Lasers clearly form the only suitable light sources. However, one cannot readily use a laser to study its own resonance radiation. This is because population inversion cannot normally be achieved when the lower state is long lived, and a long-lived state is necessary to provide sufficient population for scattering experiments. Unfortunately, an overlap of a laser line and a suitable resonance line in a different element have not yet been found.

We are presently investigating two possibilities for laser excitation of the $6^2S_{1/2} - 6^2P_{3/2}$ transition in cesium at 8521.12 Å. The first involves the recently developed semiconducting gallium-arsenide injection lasers.⁽³⁾ The emission wavelengths of these devices at liquid-nitrogen temperature usually fall in the 8450-8500-Å region, and are continuously tunable over a range of about 50 Å. We have obtained a number of these lasers and have made various measurements of their properties. Several lasers were found to be tunable well above 8500 Å, and it is clear that cw emission at 8521 Å can be obtained. The main problem associated with these lasers is wavelength stability. Because of the temperature dependence⁽⁴⁾ of the energy gap of gallium arsenide (this is the tuning mechanism), temperature fluctuations in the laser result in broadening of the emission. Success of this type of experiment depends upon careful regulation of the temperature, or working out a method to compensate for the fluctuations. One type of compensation⁽⁵⁾ is to

lock the laser to one side of the cesium line and to use the resulting error signal to control the power in the laser, which in turn determines its temperature.

The second possibility we are considering is to make use of the $4S_2 - 4P_2$ transition in argon, which falls at 8521.44 Å or 0.46 cm^{-1} below the cesium line. Although this transition has not yet been observed to lase, it appears possible to make it do so by optimizing the various parameters involved.⁽⁶⁾ If this can be done, it will provide a more intense source with a narrower linewidth than can be obtained with injection lasers. We have obtained a pair of narrow-band confocal mirrors and an interference filter in preparation for making this laser.

The argon and cesium lines can be brought into coincidence most simply by using the Zeeman effect to shift the cesium line. This method is very useful since it allows tuning the resonance line through the (fixed) laser line. We have set up apparatus to determine the magnetic field required, using a simple electrodeless rf discharge lamp and a cesium absorption cell. The spectrum of the argon lamp was measured at a resolution of 5 Å, and was found to have a pure unreversed emission line spectrum. The 8521-Å line is separated by more than 100 Å from the nearest strong lines and is comparatively strong.⁽⁷⁾ We found several strong absorption maxima below 10 kG.

Another method of utilizing the argon line results from the fact that the energy of the argon transition is less than that of the cesium. The energy difference can be made

up with microwave photons at $\lambda = 2.2$ cm, a convenient wavelength from the experimental aspect. In this case, the interaction is a double photon process.

It is expected that a cesium beam will be necessary to reduce the Doppler spread of the 8521-Å line.

Program for the next interval: Efforts will be made to make argon lase on the transition at 8521 Å. If this is successful, optimization and final design of the laser will be made. If this is not successful, methods for operating injection lasers will be further studied. It is expected that design of the scattering apparatus will be undertaken during the next interval, and unless difficulties arise with the laser, construction will begin.

(1) W. Heitler, The Quantum Theory of Radiation, 3rd ed., (Oxford University Press, 1954), p. 201.

(2) Ibid., pp. 348-353.

(3) G. Burns and M. I. Nathan, Proc. IEEE 52, 770 (1964).

(4) H. Statz and C. L. Tang, J. Appl. Phys. 36, 3923 (1965).

(5) W. E. Engeler and M. Garfinkel, J. Appl. Phys. 34, 2746 (1963).

(6) W. R. Bennett, Jr., private communication.

(7) American Institute of Physics Handbook, (McGraw-Hill Book Co., New York, 1957), p. 762.

C. COLLISION PHENOMENA

1. Optical Excitation with Low-Energy Ions*

(S. Dworesky, R. Novick, W. Smith, N. Tolk)

We have observed the optical excitation of helium in low-energy (below 500 eV) He^+ -He collisions. Three lines in the visible spectral region have been identified corresponding to the following atomic helium transitions: $2^3\text{S}-3^3\text{P}^0$, $2^3\text{P}^0-4^3\text{D}$, and $2^3\text{P}^0-3^3\text{D}$. Although very low-intensity radiation was detected from the $2^1\text{P}^0-3^1\text{D}$ transition, no other lines corresponding to singlet transitions (for example, $2^1\text{S}-3^1\text{P}^0$ and $2^1\text{P}^0-4^1\text{D}$) were observed in this preliminary study. Also, no radiation arising from S-state parent levels was found. Our results indicate that triplet states are favored over singlet states and that D and P states are strongly favored over S states. DeHeer *et al.*⁽¹⁾ found that in the capture cross sections, the maxima into D levels occur at low energies, while the corresponding P and S level maxima occur at successively higher energies. This is consistent with our observation. The results just described were obtained with the apparatus originally constructed for studying two-photon decay of metastable He^+ ions. The McPherson 0.3-m scanning monochromator discussed in the previous report⁽²⁾ was used, but not with maximum efficiency because the largest observable solid angle of the scattering chamber corresponded to approximately $f/8$. A new scattering apparatus, designed specifically for high-efficiency optical work, and a He^+ ion source for increased beam current have been developed and tested. The development of this equipment and its initial operation will subsequently be discussed.

The vacuum system of the new apparatus consists of three differentially pumped chambers (see Fig. 9). Mercury diffusion pumps topped with thermoelectric and liquid-nitrogen baffles are used throughout. The first chamber houses the source; the second chamber contains a system of electrostatic lenses which guide the ion beam from the source chamber to the third chamber in which the scattering will take place. The scattering chamber is outfitted with a window through which the emitted light can be observed and with a Faraday cup to monitor the beam. A considerable improvement over the previous apparatus was effected by making the scattering chamber much smaller. This allows photodetectors to get closer to the beam and increases the observable solid angle significantly.

A major deficiency in the old apparatus was the inability of the He^+ source to produce a sufficiently intense He^+ beam at very low energies. The long times required to accumulate adequate statistics seriously hampered our studies.

We have attempted to solve this problem by developing an entirely new He^+ ion source. The source is illustrated in Fig. 10. One major innovation is the use of an oxide-coated cathode which has the capability of emitting a greater number of electrons at lower temperatures than would a tungsten filament. The cathode surface is approximately 1 in. x 1/2 in.

The source itself consists essentially of three nested rectangular boxes. All the boxes are made of mesh or of metal drilled with numerous holes to insure good pumping and to increase transparency. The largest box is grounded; suspended, but insulated, from it is the cathode. The top of

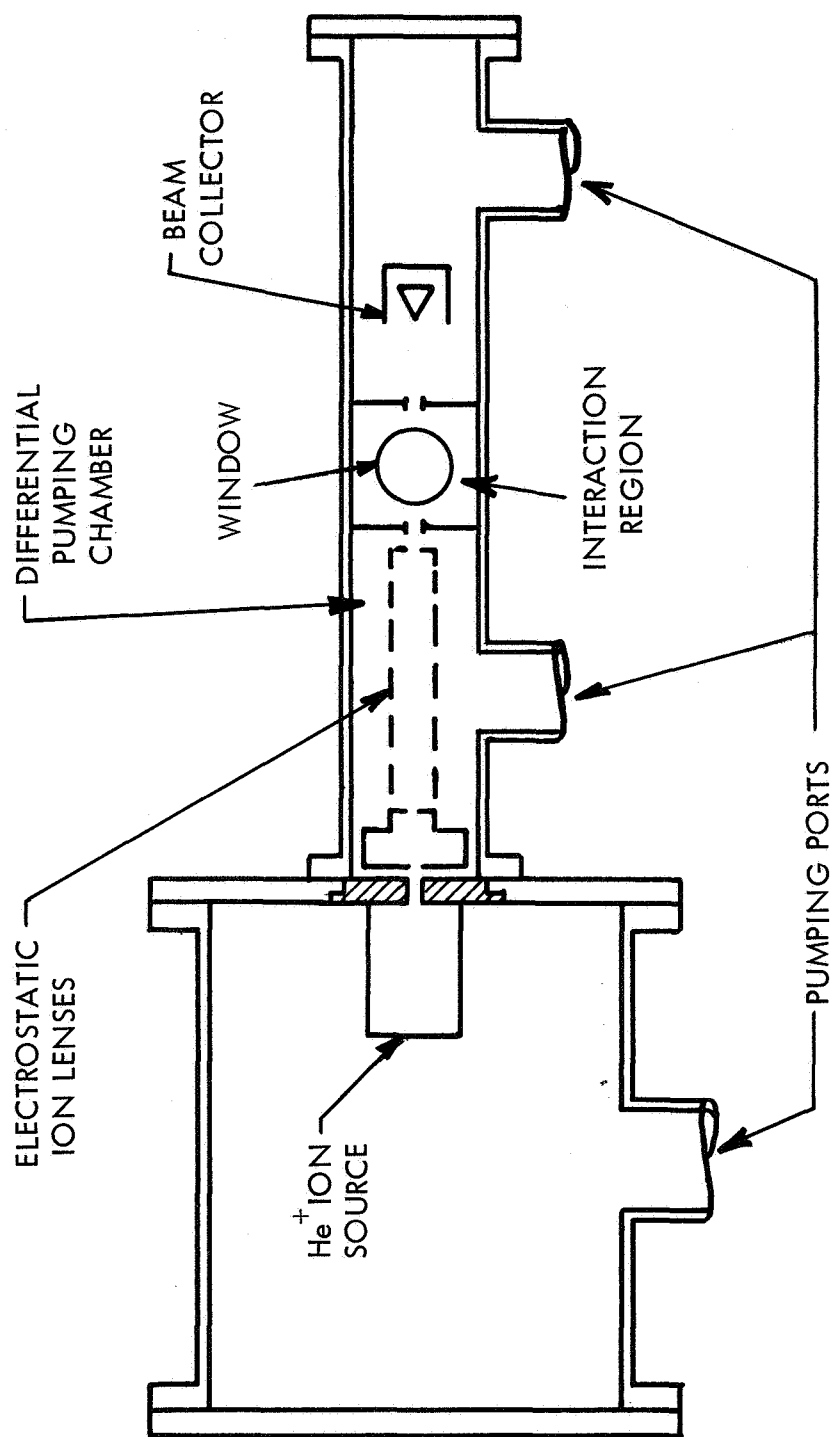


Fig. 9 Ion-atom scattering apparatus.

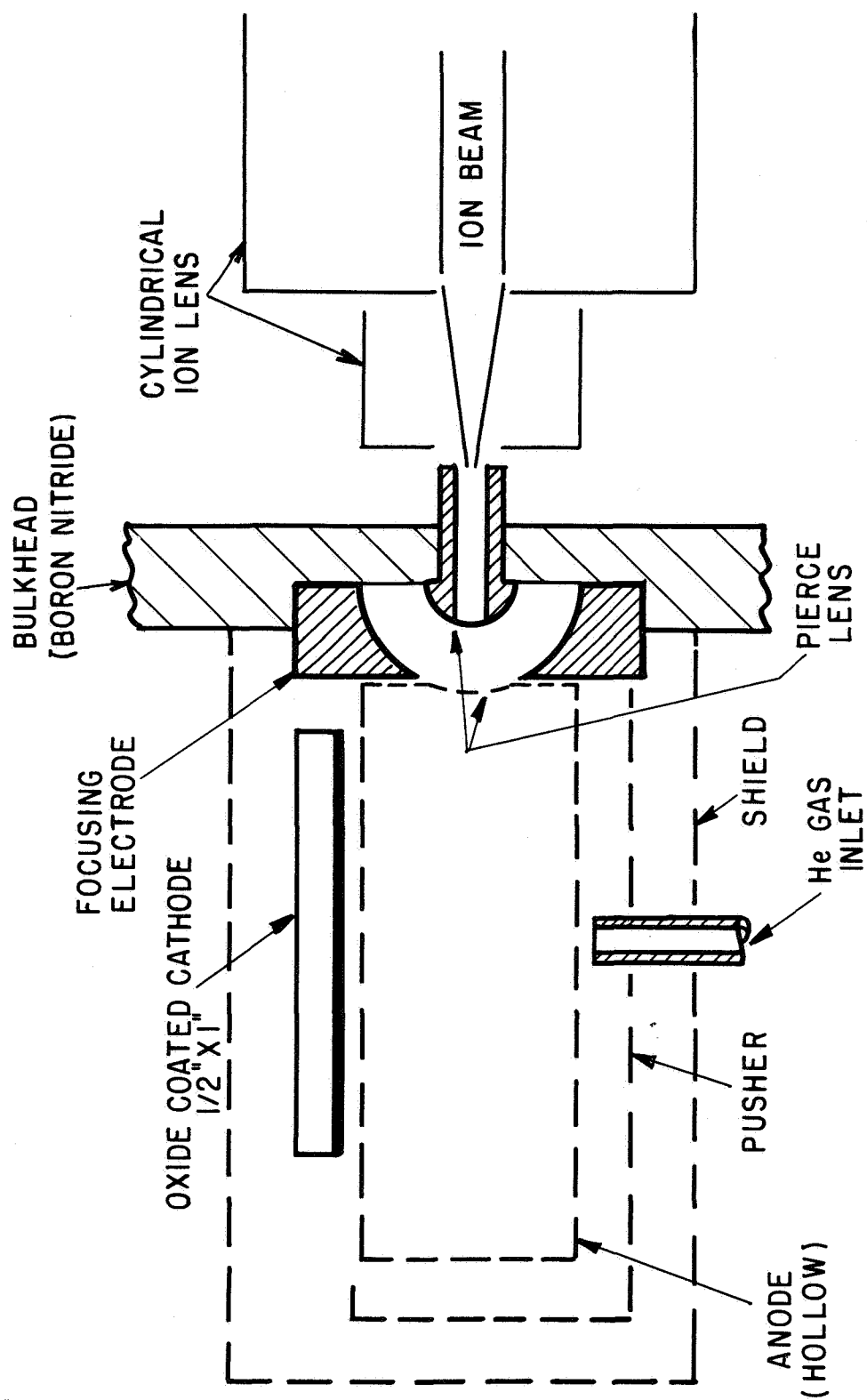


Fig. 10 He⁺ ion source.

the middle box consists of a grid of $\approx 75\%$ transparency which allows entrance of electrons into the smallest box. The smallest box (anode) is made of molybdenum mesh wrapped around a molybdenum frame. The source is designed to produce He^+ in the anode box. The He^+ is extracted as a beam through an electrostatic Pierce-type lens located at the front of the anode. Helium gas is admitted through the bottom of the source as shown. An electrostatic lens system is placed beyond the Pierce lens to guide the beam to the interaction chamber.

An effective innovation was made to increase beam current. The voltage between the Pierce lens and the anode is just the extraction voltage of the He^+ ions from the anode. The beam voltage, however, is the voltage between the anode and ground and is independent of the extraction voltage. In the previous source the extraction voltage was the beam energy. This innovation allows us to extract a much larger low-energy beam by increasing the extraction voltage on the Pierce lens without succumbing to space-charge limitations in the differential pumping channel.

A point of conjecture in the development of the source was whether the grid was doing more good by preventing the loss of He^+ back to the cathode or more harm by preventing electrons from reaching the inner box. Removal of the grid has convinced us that the gridless mode of operation is more efficient. The source consequently is now operated with the cathode directly exposed to the top face of the inner box (anode). The new spacing between the anode and cathode is the same as it was between the grid and the cathode.

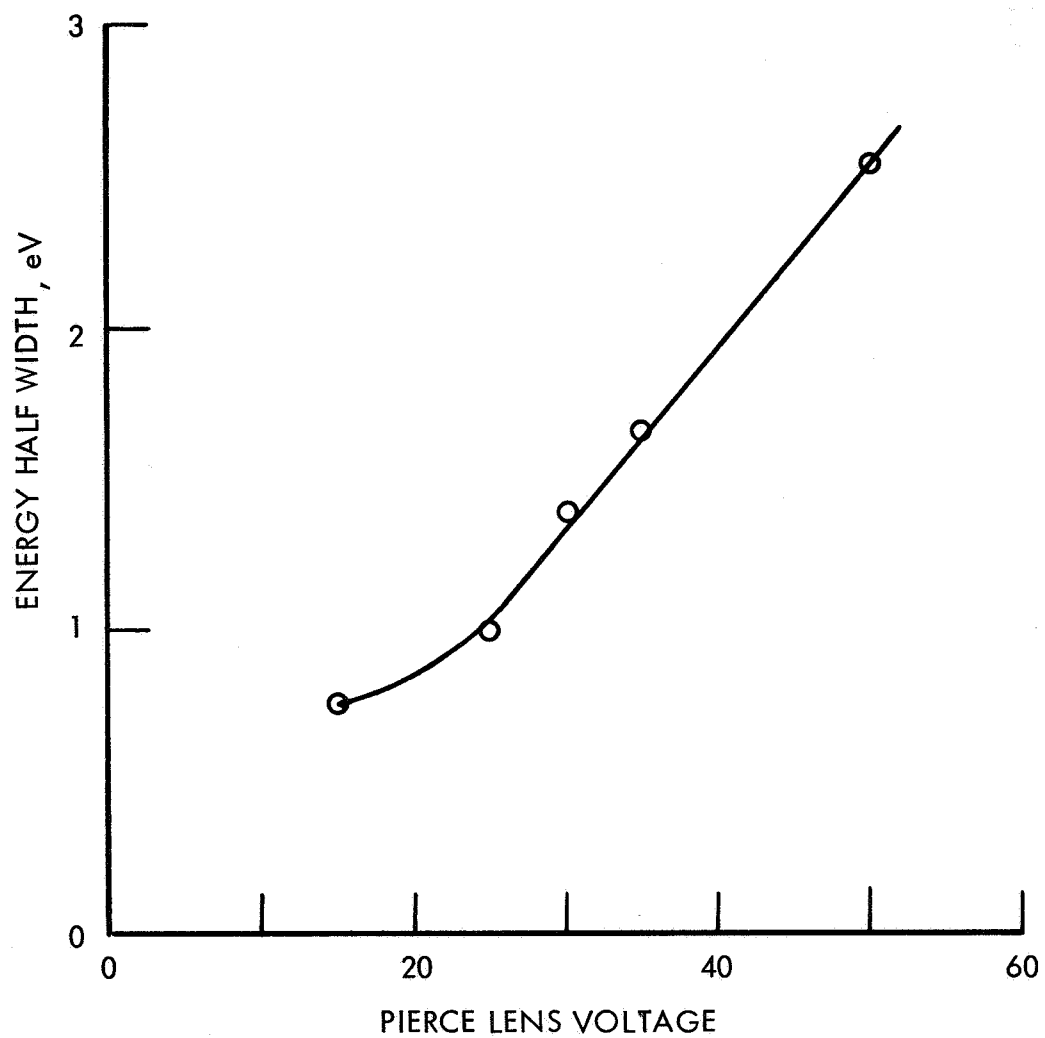


Fig. 11 Variation of the energy half-width of He^+ ion beam with Pierce-lens voltage.

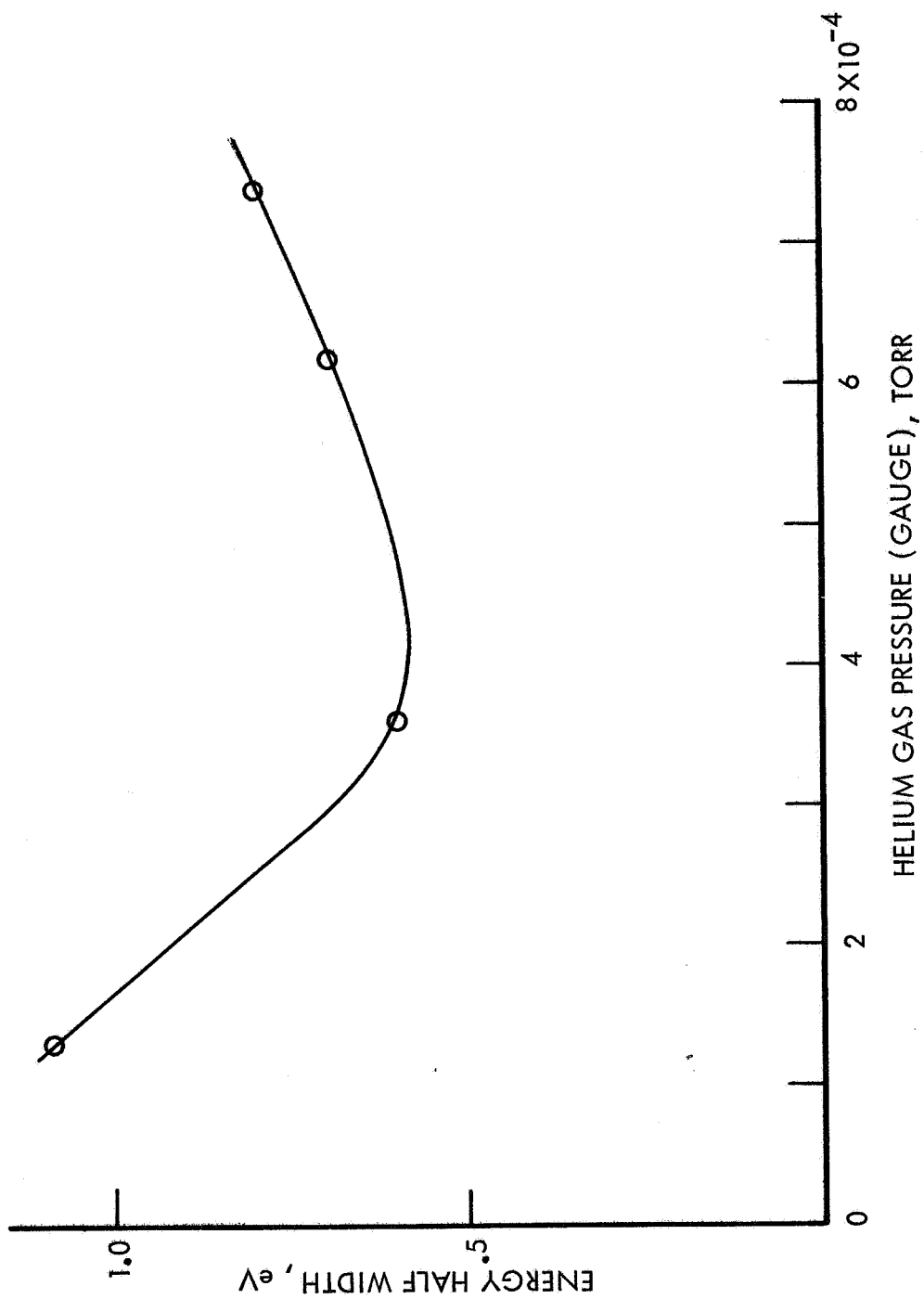


Fig. 12 Variation of the energy half-width of He^+ ion beam with helium gas pressure in the source chamber.

Optimizing all parameters, we have obtained a He^+ beam of 2×10^{-6} A. At a beam voltage as low as five volts, a beam intensity of about 3×10^{-7} A has been consistently obtainable, with an energy spread of less than one volt.

The energy half-width of the beam has been studied as a function of the various source parameters and helium gas pressure. With the exceptions of the Pierce-lens voltage and the pressure variation, the energy half-width has been found to be relatively insensitive to parameter variations. Figures 11 and 12 show the variation of energy spread with Pierce-lens voltage and with helium gas source chamber pressure.

Program for the next interval: 1) To complete development of the new ion source and scattering apparatus. 2) To identify further states involved in low-energy ion-atom scattering processes. 3) To begin a program of measurement of ion-atom collision excitation cross sections.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) F. J. DeHeer, L. Wolterbeck Muller, and R. Geballe, *Physica* 31, 1745 (1965).

(2) CRL Progress Report, April 30, 1966, p. 25.

2. Relaxation of Optically Oriented Atoms (W. Happer, E. B. Saloman)

Since the last Progress Report we have shown that it is convenient to use light modulation signals to measure orientation relaxation times. Preliminary experiments have been performed with double-resonance signals in the $(5s5p)^3P_1$ state of cadmium. A schematic diagram of the apparatus used in these experiments is shown in Fig. 13. Signals at the

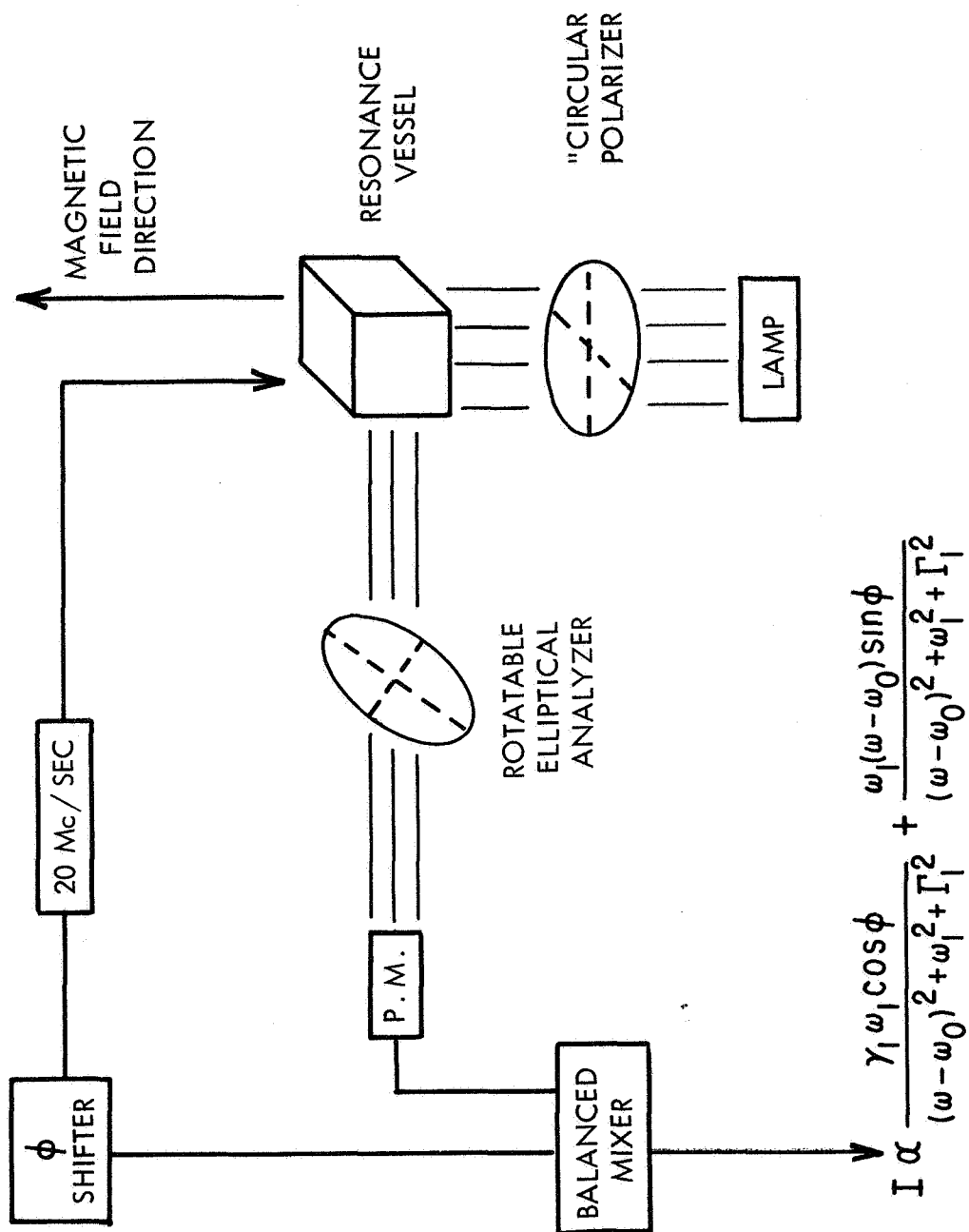


Fig. 13 Schematic diagram of apparatus to measure Γ .

driving frequency ω (20 Mc/sec) of the rf field are detected with a fast response time photomultiplier tube. A rotatable elliptical analyzer is used to null out signals from the alignment of the excited state. One of the axes of the ellipse must be parallel to the static magnetic field. This condition is easily attained empirically by removing the circular polarizer from the path of the incident light and rotating the elliptical analyzer until no signal is recorded upon sweeping the magnetic field through the resonance. When a circular polarizer is inserted into the input arm, strong signals originating purely from the orientation of the vapor are observed. These signals were detected coherently by mixing the output of the photomultiplier tube with a phase-shifted signal from the rf generator. The problem of rf pickup was avoided by chopping the light at a low frequency and detecting the output of the 20 Mc/sec balanced mixer in a conventional lock-in amplifier. Consequently, no elaborate rf shielding was required.

The long-term stability of this system proved to be exceptionally good. Once the elliptical analyzer was adjusted to null out the alignment signal, no readjustment was necessary for periods of several days. This was a considerable improvement over the Hanle-effect apparatus described previously,⁽¹⁾ where readjustments of the polarizers were necessary every 15 minutes. A further advantage of the present apparatus is that the degree of circular polarization of the incident light is not critical and fairly eccentric elliptical polarizers still give good signals.

We hope to observe relaxation induced by foreign gases with this apparatus in the near future.

(1) CRL Progress Report, June 15, 1965, p. 20.

D. ATOMIC FREQUENCY STANDARDS

1. Optically Pumped Masers (B. S. Mathur, R. Novick)

Components of the rubidium maser are being reassembled in order to resume the Stark-shift and frequency-modulation experiments. Rubidium atoms are optically pumped to the hyperfine upper ground state by Rb^{87} 7800-Å light. A Rb^{85} cell is used as a filter to eliminate the undesired $5S_{1/2}(F=2)$ hyperfine component from the pumping light. The cell can be adjusted for effective filtering by applying an electric field to shift the $6P_{3/2} - 5S_{1/2}(F=3)$ line of Rb^{85} downward in frequency to coincide with the $6P_{3/2} - 5S_{1/2}(F=2)$ line of Rb^{87} . A study will be made of the rubidium maser output power as a function of the electric field applied to the Rb^{85} filter cell. A very moderate electric field can be used to adjust the filtered Rb^{87} pumping line profile.

In the experiment for the detection of optical pumping with nonresonant light, the modulation of the off-resonant crossbeam at the Zeeman frequency of the ground state of rubidium has been observed.⁽¹⁾ Extension will be made of these experiments to look for the modulation of the pumping light at the ground-state hyperfine frequency of rubidium.

A scanning Fabry-Perot interferometer is being used to study the effect of an electric field on the ground-state hyperfine separation of rubidium. Because of the small-diameter reflecting mirrors and the lack of sensitive detecting methods, resolution of the rubidium hyperfine structure has not been successful. A further attempt will be made with

larger-diameter mirrors and solar cells with low-noise amplifiers which can detect low power for alignment of the scanning interferometer. An improvement in the optics of detection will also be tried.

Program for the next interval: To complete the assembling of the rubidium maser; to attempt again resolution of the rubidium ground-state hyperfine structure; to make preliminary Stark-shift measurements; to try to observe the modulation of the pumping light at the microwave frequency of the maser transition.

(1) CRL Progress Report, October 31, 1966, p. 45.

2. Detection of Optical Pumping with Nonresonant Light (W. Happer, B. S. Mathur)

An experiment to detect the modulation of nonresonant light at the ground-state Zeeman frequency of rubidium was described in the previous Progress Report.⁽¹⁾ During this period the detection technique was improved and modulation was observed.

The experimental arrangement (Fig. 14) is very similar to that of the original crossed-beam experiment suggested by Dehmelt.⁽²⁾ Linearly polarized Sr-II light from a microwave excited flow lamp passes through an optically pumped Rb vapor contained in a 300-ml Paraflint-coated spherical flask. Interference filters are used to isolate the appropriate optical lines. A transverse oscillating magnetization is induced in the Rb vapor with an rf magnetic field at the Zeeman frequency. The vapor then behaves like an optically active crystal which rotates the plane-polarized light either to the right or to the left depending

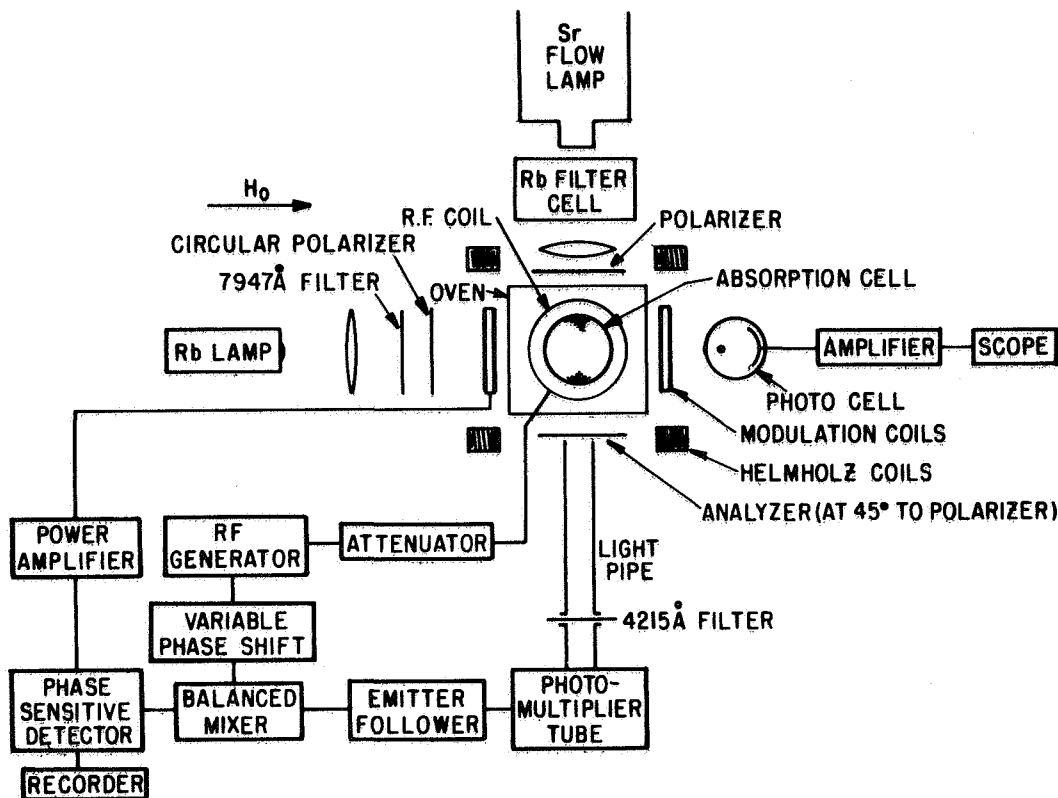


Fig. 14 A schematic diagram of the apparatus.

on the direction of the magnetization. After traversing the Rb vapor, the probing light passes through a linear polarizer whose axis is rotated by 45° to the initial polarization axis of the probing beam. The light transmitted by this polarizer is intensity modulated at the Zeeman frequency. The modulated light is detected by a photomultiplier tube and associated electronics. A typical Zeeman resonance detected with

Sr-II probing light is shown in Fig. 15. The bandwidth of the detection system was 1 cps.

Zeeman resonances in both Rb^{85} and Rb^{87} were observed in this way. The Rb^{85} signals were about twice as large as the Rb^{87} resonance; this is to be expected since one of the Rb^{85} hfs components is very close to the Sr-II line.⁽³⁾

The Sr-II light was filtered with a heated natural rubidium filter cell in order to absorb any of the Sr-II lamp profile which coincided with the Rb absorption profile. The Zeeman resonances in the transmitted rubidium pumping light were monitored with a silicon photodiode. No change in the amplitude of these resonances was observed as the Sr-II probing light was turned on and off. Furthermore, the two linear polarizers were removed from the path of the probing beam, and, just as in the original crossed-beam experiments,⁽²⁾ circularly polarized Sr-II light was used as a probe. No Zeeman modulation could be detected on the probing beam, even when the natural Rb filter cell was removed from the path of the Sr-II light. We conclude that no detectable absorption of the probing beam occurred, and that the populations of the rubidium ground-state sublevels were not affected by the Sr-II light.

Rubidium lamps were also used to supply the probing beam. No modulated signal was observed with the 4215-Å resonance line even though an intense flow-lamp source was used. This is to be expected if the D_1 and D_2 lines have approximately equal intensities. The signal from the 7800-Å Rb light was recorded as a function of the temperature of the filter cell. An interesting reversal of the phase of the

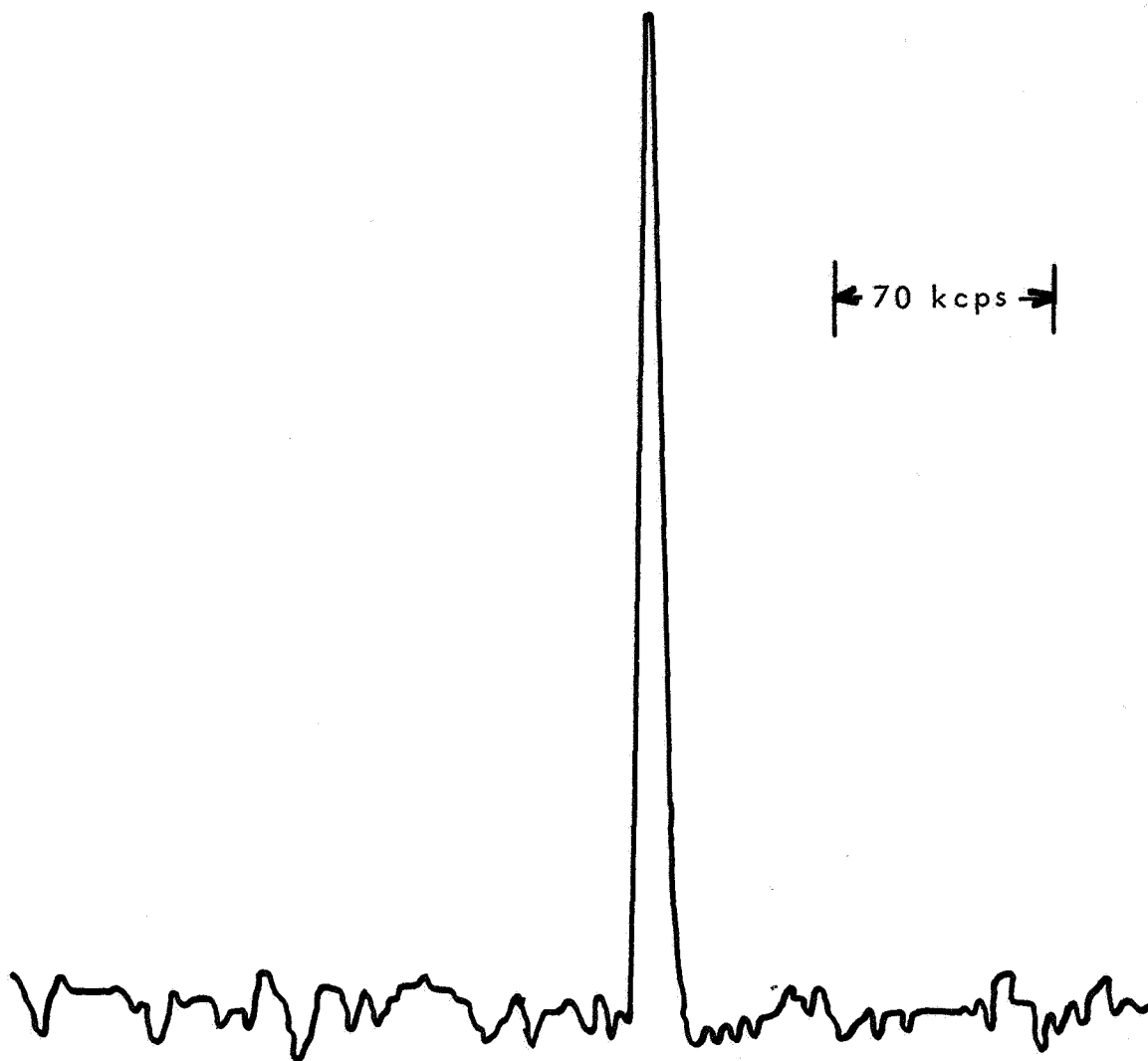


Fig. 15 A recorder trace of the phase-sensitive detector output as the magnetic field is swept through the resonance. The phase-sensitive detector time constant was 1 sec.

signal was observed at a temperature of about 75°C. We were unable to see any signal at temperatures above 160°C. The cause of this phase reversal is not yet clear, but it is probably related to the change in the spectral distribution of the probing light.

Program for the next interval: (1) To make a cylindrical absorption cell with which to study the pumping as a function of the distance from the pumping end, using the Sr-II line as the nonperturbing probe; (2) to use different buffer-gas-filled absorption cells, and to try to understand the phase-reversal phenomenon; (3) to work out a theory for the above experiment.

(1) CRL Progress Report, April 30, 1966, p. 34.

(2) H. G. Dehmelt, Phys. Rev. 105, 1924 (1957); W. E. Bloom and A. L. Bloom, Phys. Rev. 107, 1559 (1957).

(3) J. Winocur and R. V. Pyle, J. Appl. Phys. 36, 2740 (1965).

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II.3 PHYSICS OF MOLECULES 81010

A. BEAM MASER SPECTROSCOPY*
 (P. Cahill)

An analysis of the observed spectra of NHD_2 and NH_2D was made with a hyperfine Hamiltonian⁽¹⁾ which contained quadrupole and spin-rotation interaction terms for the nitrogen and deuterium nuclei in addition to spin-spin interactions for the four nuclei. The coupling constants for the four spin-spin interactions can be calculated to a higher precision from the geometry of the particular molecule than they can be determined experimentally with the present resolution. The following vector coupling system was used to interpret theoretically the hfs.

$$\begin{aligned} J + I_N &= F_1, \\ F_1 + I_D &= F_2, \\ F_2 + I_H &= F, \end{aligned} \quad (1)$$

where J is the rotational quantum number, and I_N , I_D , and I_H are the respective nuclei spins. An analysis⁽²⁾ had previously been made of the $3_{03}-3_{13}$ transition at 18 GHz in NH_2D with $I_H = 1$. Subsequently, the hfs spectrum of the $3_{03}-3_{13}$ transition at 43 GHz in NH_2D with $I_H = 0$ was observed in order to determine more precisely the deuterium coupling constants. For NHD_2 , measurements have been made of the $3_{03}-3_{13}$ and $2_{02}-2_{12}$ transitions at 18 and 38 GHz, respectively, where, because of the symmetry of the molecular states, $I_D = 1$ in the vector coupling system (Eq.1). In all three transitions, the coupling constants are calculable for

the nitrogen nucleus as well as for the spin-spin interactions. Initially, the matrix elements for the Hamiltonian were calculated using the available tables of Wigner 6j coefficients. Since this calculation for the two hfs transitions of NH_2D consumed an enormous amount of time, a computer program⁽³⁾ was obtained and modified for Fortran 4. All the matrix elements for the quadrupole, spin-rotation, and spin-spin interactions in the vector coupling scheme were calculated with the use of the 7094 computer. Also, the hfs frequencies and intensities for transitions corresponding to $\Delta F_1 = 0$ were calculated for various values of the coupling constants. The observed spectrum was plotted with a General Dynamics SC4020 plotter assuming a 5-kHz Gaussian line shape for each transition. A direct comparison is then obtained between the calculated and observed hfs spectrum. The 43-GHz transition, which involved only five coupling constants, was analyzed first. Figure 16 shows the calculated spectra for NH_2D . To obtain the best fit of the observed spectrum, a graphical analysis was made by plotting the deuterium quadrupole constants for the upper and lower states versus the frequency difference between two particular hfs transitions. This is shown in Fig. 17 for the frequency separation between the observed lines 4 and 5. The observed and calculated frequencies for the best fit for the coupling parameters is given in Table III. Although in the calculated spectrum the three most intense lines corresponding to the nitrogen hfs transitions are shown to be split by approximately 8 kHz, no splittings were actually observed. The variation between the observed and calculated lines is attributable in part to a

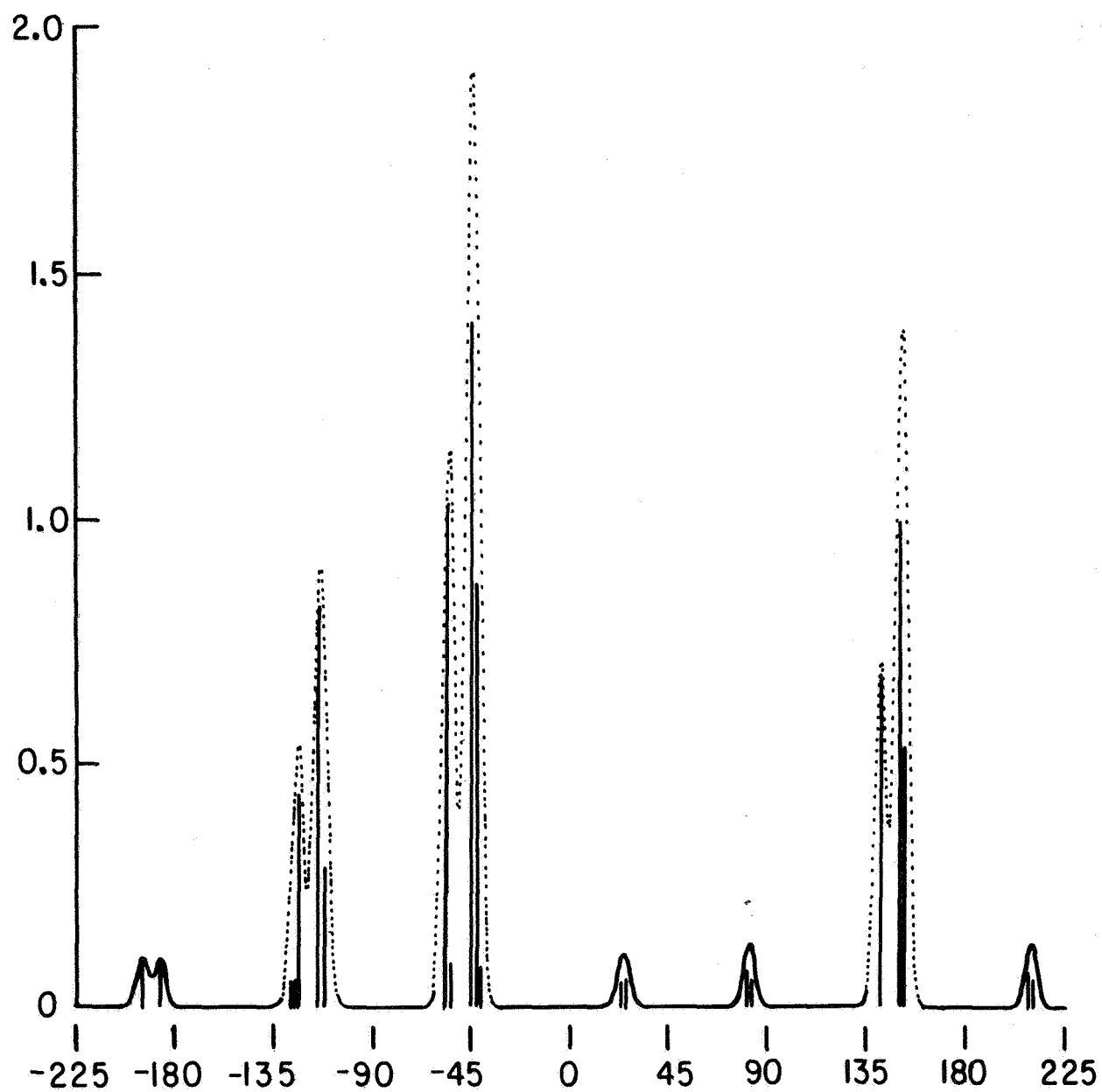


Fig. 16 Calculated spectra for NH_2D .

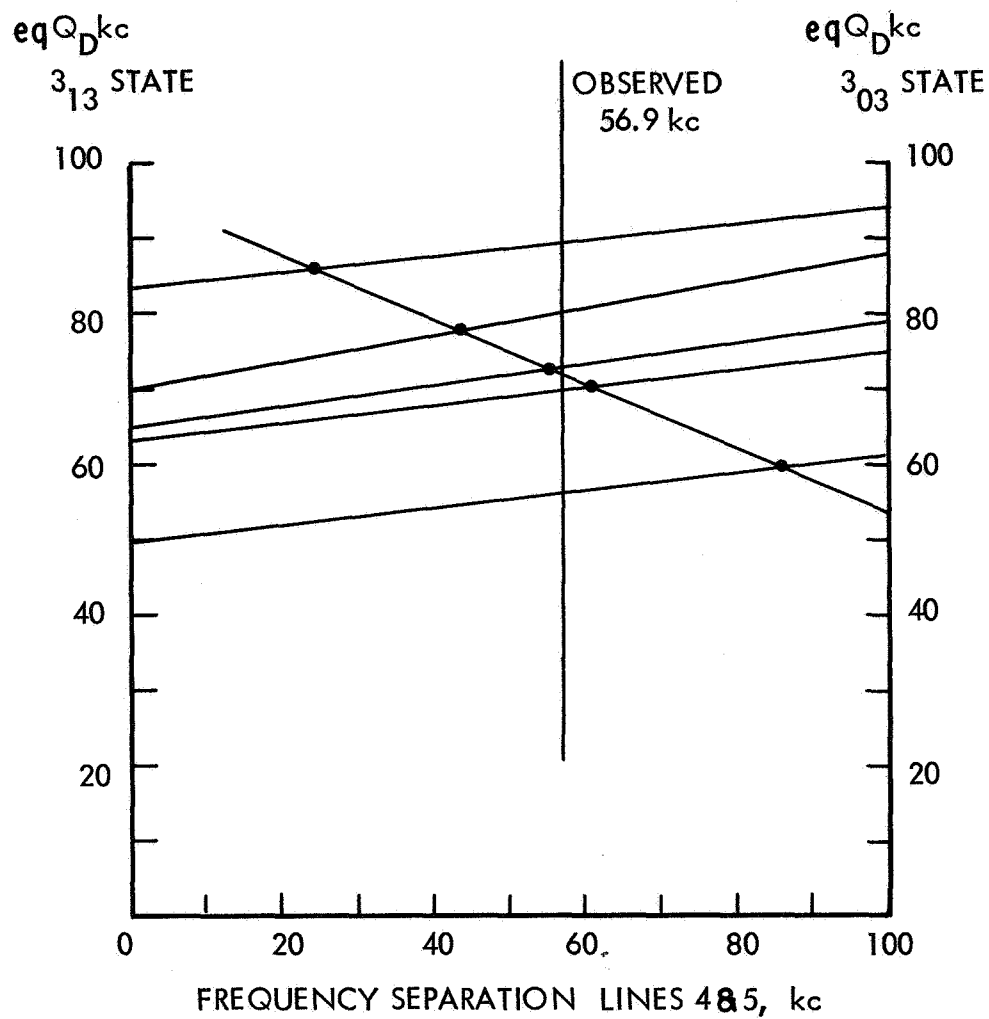


Fig. 17 Plot of frequency separation between lines 4 and 5 in NH_2D as a function of deuterium quadrupole moments for the two states.

TABLE III

Calculated Frequencies of NH_2D at 43.1 GHz.

Observed (kHz)	Calculated (kHz)	Coupling Constants (kHz)		
		States		
		<u>3 13</u>	<u>3 03</u>	
-138.1	-137.7	-63.0	-75.0	(eqQ)D
- 71.5	- 68.1	10.0	10.0	CN
0	0	- 2.7	- 2.7	CD
66.3	69.8	-1566.5	-1369.3	(eqQ)N
123.2	129.2	0.50	0.70	μ spin-spin (D•N)
192.1	191.5			
246.9	249.9			

reading error of the receiver used in the measurements and in part to inadequate spectral resolution. For the two observed transitions in NHD_2 , the frequency of the receiver's oscillator was measured directly. Presently, the agreement between observed and calculated frequencies is approximately 4 kHz, which corresponds to a measurement accuracy of better than one part in 10^7 . In order to observe the variations of the deuterium spin-rotation constant in the different states, a measurement of the observed spectral lines to at least five parts in 10^9 would be required. Because of the interest in comparing theoretical and observed values of the spin-rotation constants as a function of rotational state, the spectra of NH_2D at 43 GHz is in the process of being re-examined with

continuous counting of the receiver's frequency. Also to do a more refined treatment of the data, a variation of the deuterium quadrupole and spin-rotation coupling constants should be performed using as a criterion of minimization the variance between the calculated and observed frequencies. A general least-squares program is being written.

For the hfs spectra for the $2_{02}-2_{12}$ transition in NHD_2 , an agreement between observed and calculated frequencies to at least 4 kHz is currently attained. A final assignment will partly depend on the hfs constants determined for NH_2D .

In this period attempts were also made to observe the hfs spectra of the $5_{23}-6_{16}$ transition in H_2O using a TM_{020} mode cavity. The spectrum was not observed under the usual operating conditions, nor when the detection efficiency was improved by modulation of the focusing voltage. Finally, a TMC Nemotron was used to sum several sweeps over the frequency range of the cavity. No lines were observed. Effectively, because of the high J states of the molecule, electrostatic focusing of the individual states decreases too rapidly with J to obtain any appreciable population difference between the two states. Also a TM_{020} mode cavity was constructed for the $1_{11}-1_{10}$ transition in HDS at 51 GHz. The current objective is to measure the rotational dependence of the spin-rotation interaction constant in HDS.

Program for the next interval: The 43-GHz transition of NH_2D will be examined, and the assignment of the spectra for the deuterated ammonias will be completed and prepared for publication. The hfs of HDS will also be examined at 51 GHz.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-66 and in part by the Office of Naval Research under Contract N00014-67-A-0108-0002.

(1) P. Thaddeus, L. C. Krisher, and J. H. N. Loubser, J. Chem. Phys. 40, 257 (1964).

(2) P. Thaddeus, L. C. Krisher, and P. Cahill, J. Chem. Phys. 41, 1542 (1964).

(3) M. H. Pine (private communication).

B. ELECTRON EXCITATION OF MOLECULAR HYDROGEN*
(P. Cahill)

During the last few years several experimental investigations have been carried out on the polarization of atomic line radiation excited by electron impact. For the first time we report on a similar process for the excitation of the $^3\Pi_u(1s3p)$ state of H_2 in a single collision process by electrons. The hydrogen emission line, $\lambda = 6226 \text{ \AA}$, corresponding to an $N = 2$, Q branch ($N =$ total angular momentum apart from spin) transition from the $^3\Pi_u$ to $^3\Sigma_g(1s2s)$ states in orthohydrogen, was excited in a gas-filled tube by a parallel electron beam. The electron gun used consisted of an oxide-coated cathode, two accelerator grids, and an anode at the same potential as the second grid. This particular transition was separated from the other lines of atomic and molecular hydrogen by means of a filter, which had a width of $10 \text{ \AA}$ at half transmission. The radiation intensities $P_{||}$ and $P_{\perp}$ polarized parallel and perpendicular, respectively, to the electron beam were measured as functions of the electron energy. The light was passed through a mechanically

rotating polaroid, allowing $P_{\parallel} - P_{\perp}$ and $P_{\parallel} + P_{\perp}$ to be measured and compared simultaneously. A diagram of the apparatus is given in Fig. 18.

Figure 19 shows the measured polarization and excitation cross section as functions of electron energy. The maximum polarization is observed just above threshold. Because of the poor signal-to-noise ratio, the minimum voltage at which polarization could be observed was 18 V, which is approximately three volts above the theoretical excitation threshold. The line intensity of the excited hydrogen line was proportional to the gas pressure as well as to the electron beam current in those ranges quoted in the text of Fig. 19.

A generalized expression describing the magnetic hyperfine interactions in a homonuclear diatomic molecule has been obtained for the case of Hund's coupling $b\beta_J$. The effect of the magnetic hyperfine interactions on the fine structure has been calculated for the $c^3\Pi_u(1s2p)$ state of molecular hydrogen. Using Chiu's wave function, we obtained the following results for the coupling constants:⁽¹⁾ $a = 20.1$ MHz, $b = 1096.2$ MHz, $c = -8.9$ MHz, and $d = 89.7$ MHz. These results, which differ from those of a previous calculation, are being prepared for publication.

Program for the next interval: Further experiments will be done on the measurement of the polarization of H_2 as a function of pressure and magnetic field. Also calculations on the theoretical polarization for the excitation of a diatomic molecule will be completed, and hyperfine coupling constants for the $^3\Pi_u(1s3p)$ state of orthohydrogen will be calculated.

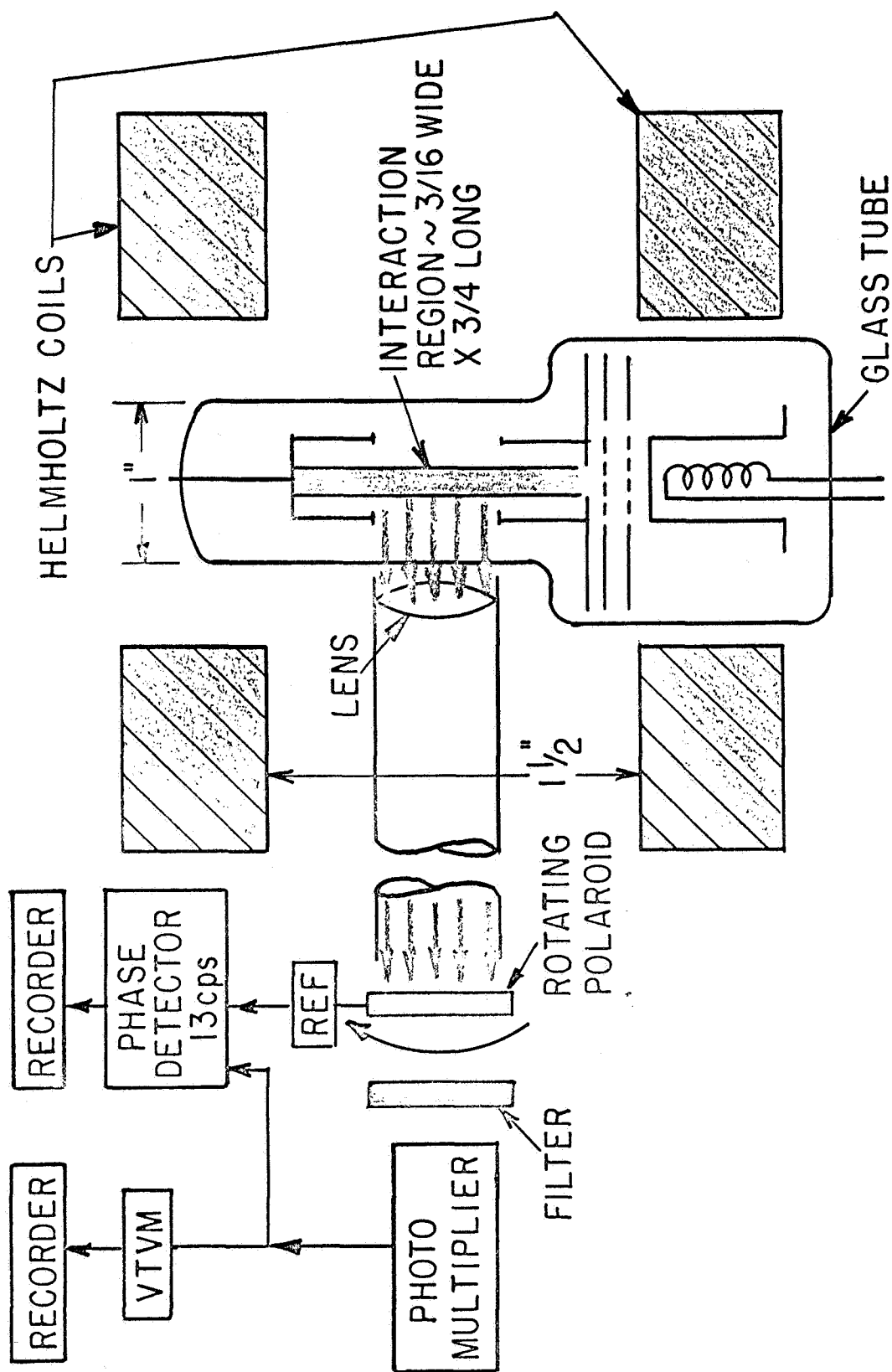


Fig. 18 Schematic diagram of apparatus.

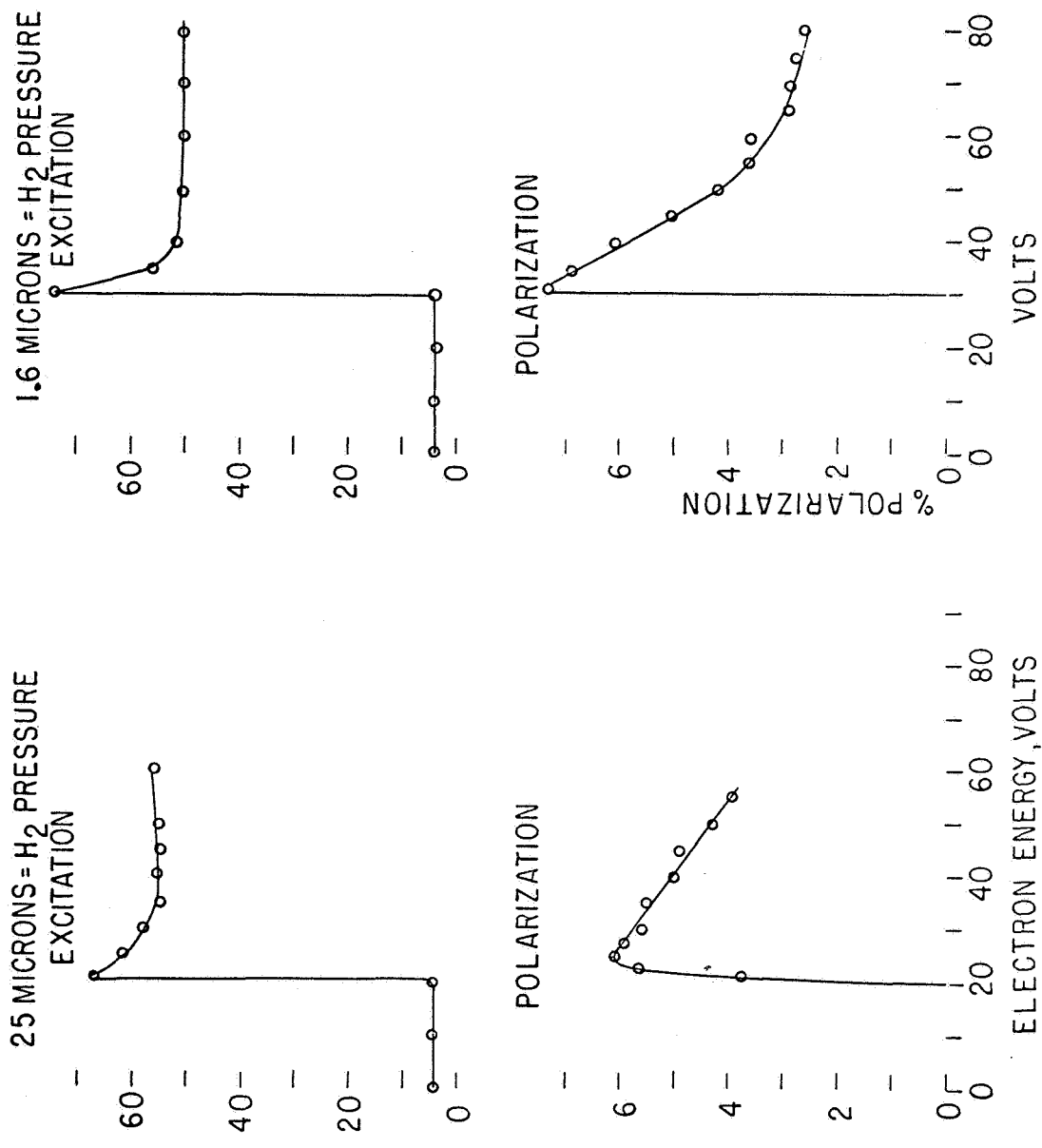


Fig. 19 Polarization and excitation curves.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-66 and in part by the Office of Naval Research under Contract N00014-67-A-0108-0002.

(1) Notation is consistent with that used in C. H. Townes and A. L. Schawlow, Microwave Spectroscopy (McGraw-Hill Book Co., New York, 1955).

C. SCATTERING OF VELOCITY SELECTED MOLECULAR BEAMS* (P. Cahill, S. Skwire)

The experimentally measured total cross section is given approximately by the expression

$$\sigma = 2\pi \int_{\alpha}^{\pi} \frac{d\sigma(\theta)}{d\Omega} \sin \theta d\theta ,$$

where α is the resolution angle of the detector. Because α , whose value depends only on experimental geometry, is finite, some beam particles which have undergone small deflections are detected. Consequently, the measured beam intensity is greater and the corresponding cross sections are lower than would be the case for an ideal detector ($\alpha = 0$).

The theoretical cross section for the scattering of potassium by argon was corrected for the finite angular resolution of the detector.⁽¹⁾ The log of the corrected cross section had an almost linear dependence on the log of the velocity, and the slope of this line is shown as a function of α in Fig. 20. The importance of the angular resolution correction in interpreting the experimental data is clear.

In order to measure the magnitude and velocity dependence of σ as a function of α , a new apparatus having a continuously variable angular resolution is being constructed.

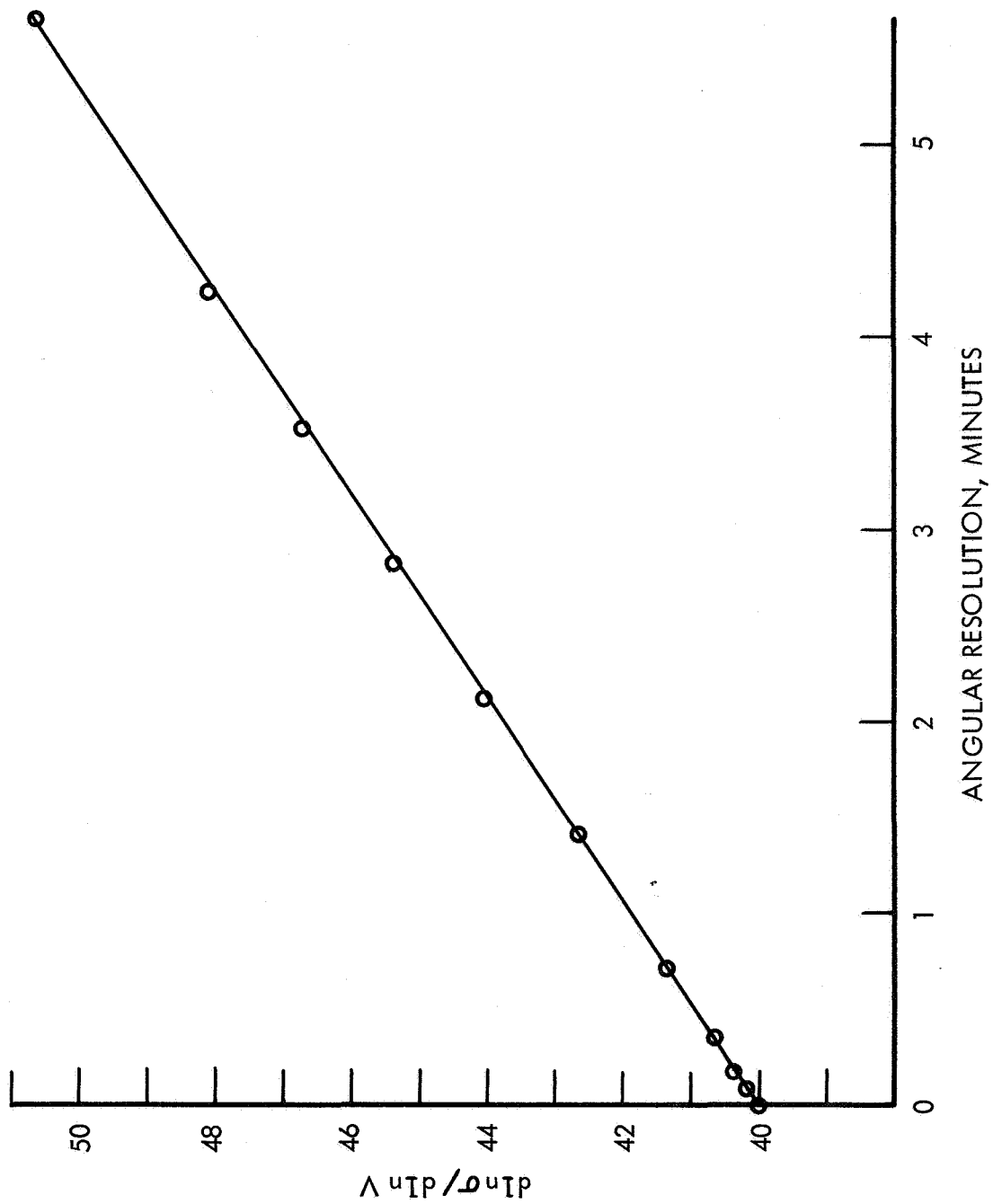


Fig. 20 Resolution correction calculated for K-Ar.

In this apparatus the distance between the detector and the scattering chamber can be varied from 1/2 to 18 in. corresponding to a change in angular resolution from 0.8 to about 15 minutes of arc, depending on the geometry of the scattering chamber. The finite size of the scattering chamber was taken into consideration in the calculation of the angular resolution. Also, the values of α , given by the expressions derived by Kusch⁽²⁾ for an infinitesimal scattering region, agreed to within six percent with the results of this calculation.

Before taking further data it will be necessary to eliminate mechanical vibrations which have developed in the detector. To this end the velocity selector motor has been replaced and further repair work has been done on the velocity selector rotor. In particular, the drive shaft has been replaced and the gears readjusted.

Program for the next interval: The velocity selector will be remounted in the vacuum system, and the effect of any remaining vibrations will be reduced by mechanically decoupling the vacuum system containing the detector from that containing the velocity selector. Also, the new apparatus will be assembled. The velocity dependence of σ for the system K-Ar and K-N₂ will be measured in order to clarify the discrepancies described in the last report.⁽³⁾

*This research was also supported by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-66 and in part by the Office of Naval Research under Contract N00014-67-A-0108-0002.

(1) R. J. Cross, Jr., E. A. Gislason, and D. R. Herschbach, J. Chem. Phys. 45, 3582 (1966).

(2) P. Kusch, J. Chem. Phys. 40, 1 (1964).

(3) CRL Progress Report, April 30, 1966, p. 38.

D. MAGNETIC ROTATION SPECTRA*

(P. Kusch, B. Palatnick)

The magnetic rotation spectrum of CS_2 has been re-photographed on the Jarrell-Ash spectrograph in order to obtain higher dispersion and resolution of the data.⁽¹⁾

While our primary interest has been in making a vibrational analysis, the new data also show resolved rotational structure, and this too may prove interesting to explore.

Extensive microphotometer traces of the new data have been taken on the Applied Research Laboratories Spectroline Scanner. The measured wave numbers of the spectral lines have been recorded on the traces which are presently being examined in the hope of discovering regularities in the very complex spectrum.

Program for the next interval: To continue to search for regularities on the traces.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-66 and in part by the Office of Naval Research under Contract N00014-67-A-0108-0002.

(1) CRL Progress Report, April 30, 1966, p. 40.

E. MOLECULAR SPECTRA OF CESIUM*

(M. M. Hessel, P. Kusch)

There is an exceedingly complex absorption spectrum of Cs_2 in the region from 6250 Å to 6600 Å. The most conspicuous features of the spectrum are the heads of the bands, degraded to the red. The rate of decrease of intensity from the band head to lower frequency is extremely variable among the bands. The heads themselves are sharp, and the frequency

of a head may easily be measured to within a few hundredths of a wave number. It has not been possible to arrange a significant number of these heads into the conventional array (Deslandres table), where the differences between the frequencies of heads in adjacent rows (or columns) are reasonably constant.

A number of discontinuities occur in which the absorption intensity drops to a low value on the long wavelength end of a region of rather constant absorption. These discontinuities do not have the rapid change of intensity near the discontinuity that is usually found in a band head. It has been possible to arrange about one hundred of these objects into a conventional square array, where the ground-state vibrational intervals are identical to those previously found in another (7500 Å) absorption system of Cs_2 . The array breaks off sharply at some value of the upper-state vibrational quantum number, and it is believed that the upper state is strongly perturbed at that vibrational level.

A large number of narrow regions occur with an intensity that is greater than would be expected from the accidental superposition of rotational lines. Rotational structure is only partially resolved even at very high resolution and dispersion. In particular, it is easily visible in the intermediate part of the spectrum and virtually unobservable at the long and short wavelength ends. This suggests a rapid change of rotational constants with vibrational quantum number. There is some evidence that the blue edges occur at the band origin in a Q branch, and that at least one

set of the red heads corresponds to a head on these Q branches with a highly variable displacement from the origin.

As an aid to the analysis of this spectrum, we have observed the fluorescence spectra of Cs_2 on a high-dispersion and high-resolution grating spectrograph. Neon has a number of strong atomic lines in the region $6250 \text{ \AA} - 6600 \text{ \AA}$, and some of these lines excited a fluorescence spectrum. The most intense fluorescence lines form a well-defined series of single lines. There is no evidence that these lines are unresolved doublets.

In Table IV we list the wave numbers, ν , of the lines of this series, the spacing, $\Delta\nu$, between adjacent lines, the second differences, $\Delta^2\nu$, and the relative intensities of the fluorescence lines. The $6402\text{-}\text{\AA}$ neon line lies in the center of this series and is the exciting line. Also listed in this table are the first and second differences, $\Delta\nu_A$ and $\Delta^2\nu_A$, respectively, of the ground state of cesium as obtained from the analysis of "blue degraded" bands in the $6250\text{-}\text{\AA}$ system.

The appearance of a series of single lines indicates⁽¹⁾ a Q-branch transition ($\Delta J = 0$). R or P transitions would give rise to a series of doublets whose components are of equal intensity.

The fluorescence data in Table IV have some anomalies. The first differences in either fluorescence or absorption are the vibrational intervals of the ground electronic state and should converge, over a modest range of vibrational quantum numbers, at a constant rate ($\Delta^2\nu = \text{constant}$) toward lower wave numbers (dissociation of the molecule). It is

TABLE IV
Fluorescence Series of Molecular Cesium Excited
by the 6402-Å Neon Line

No. of Measurements of each line n	(a) ν (cm ⁻¹)	$\Delta\nu$ (cm ⁻¹)	$\Delta^2\nu$ (cm ⁻¹)	Relative Intensity of Line	$\Delta\nu_A$ (c) (cm ⁻¹)	$\Delta^2\nu_A$ (cm ⁻¹)
7	15 972.13±0.04	40.35	0.06	5	40.33	0.18
5	15 931.78±0.03			7	40.15	
3	15 891.49±0.05	39.94	0.35	2	39.97	0.18
6	15 851.55±0.05	39.84	0.10	15	39.82	0.15
7	15 811.71±0.05	40.14	-0.30	15	39.60	0.12
9	15 771.57±0.09	39.43	0.71	2	39.47	0.13
10	15 732.14±0.02	38.87	0.56	8	39.25	0.12
10	15 693.27±0.07	39.13	-0.26	15	39.10	0.15
10	15 654.14±0.05	38.93	0.20	20	38.93	0.17
-	15 615.21	38.77	0.16	*	38.76	0.17
10	15 576.44±0.03	38.57	0.20	20	38.59	0.17
10	15 537.87±0.04	38.44	0.13	19	38.47	0.12
9	15 499.43±0.06	38.22	0.22	19	38.22	0.25
10	15 461.21±0.05	38.07	0.15	15	38.00	0.22
10	15 423.14±0.05	37.85	0.22	12		
9	15 385.29±0.06	37.72	0.13	10		
9	15 347.57±0.04	37.35	0.37	9		
5	15 310.22±0.10	37.45	-0.10	9		
4	15 272.77±0.14	36.95	0.50	3		
4	15 235.82±0.17			0		

(a) Lines with intensities greater than 3 have been measured by three different observers on four plates taken at different experimental conditions (temperature and pressure of cesium).

(b) Average error $\Sigma(\nu - \bar{\nu})/n$.

(c) These numbers come from the analysis of the blue degraded bands of the absorption spectra. Error in these intervals may be as large as ± 0.10 cm⁻¹.

* Exciting line 6402-Å neon.

true for the absorption data where $\Delta^2\nu \approx 0.17 \text{ cm}^{-1}$ within experimental error. In part of the fluorescence series from 15,347 to 15,693 cm^{-1} , there is a systematic alternation of second differences ($\Delta^2\nu = 0.13, 0.22, 0.15 \dots$). This alternation is beyond experimental error and is observed in each individual plate measurement. In other regions there is a more random irregularity in the second difference $\Delta^2\nu$. This suggests that there is a perturbation of the rotational levels of the $^1\Sigma$ ground state of Cs_2 . The nature of this perturbation is not understood.

Program for the next interval: To continue the analysis of the fluorescence data and absorption spectra of molecular cesium.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-66 and in part by the Office of Naval Research under Contract N00014-67-A-0108-0002.

(1) G. Herzberg, Spectra of Diatomic Molecules, 2nd ed., (D. Van Nostrand, Inc., New York, 1950) p. 210.

F. LASER STUDIES OF MOLECULAR BIREFRINGENCE AND OPTICAL ACTIVITY*

1. Birefringence Induced in Fluids by an Inhomogeneous Electric Field (R. L. Disch, S. Golub)

An analysis has been made of the results of the measurement of the birefringence induced by an inhomogeneous electric field in liquid benzene, and a similar analysis is in progress for carbon disulfide. The purpose of the liquid phase measurements is the determination of the sign and magnitude of the molecular electric quadrupole moment and

quadrupole polarizability under conditions of much greater signal-to-noise ratio than that attainable with gas phase measurements in most instances. This also permits a more accurate study of temperature dependence of the induced birefringence, which is essential for a complete analysis. An accurate analysis of the results of liquid phase measurements, however, depends upon an assessment of molecular correlation and distortion effects, for which no rigorous theoretical treatment is available. Fortunately, the relevant statistical mechanical average describing the correlation as well as an indication of the effective optical anisotropy of the molecule can be obtained experimentally from the Kerr effect and the depolarization ratios of Rayleigh scattered light. Further theoretical difficulties are encountered in the meaning of the Lorentz-Lorenz relation in dense anisotropic media, and we have found that the proposal of Vuks⁽¹⁾ gives more consistent results than those obtained by the use of the classical form of this relation. A publication is being prepared giving the complete analysis of the results of these measurements.

Further work in this area comprises measurements upon liquefied gases.

2. A Laser Polarimeter

(R. L. Disch, J. Matuska, D. Sverdlik)

The development of an ultra-sensitive laser polarimeter has been carried out in an effort to determine and to eliminate those factors which would prevent the attainment of its ultimate sensitivity and limit its absolute accuracy.

Virtually all polarimetric devices to date have attained sensitivities far worse than their theoretical limits. A number of optical and mechanical effects, as well as stray magnetic fields, have been shown to limit both sensitivity and accuracy, and in attempting a systematic elimination of these, we have attained a sensitivity which is within an order of magnitude of that which could be expected on theoretical grounds for the laser source employed, and some further improvement appears feasible. The present device is sensitive to an optical rotation of slightly less than 10^{-5} degree. This sensitivity permits the study of several very feeble optical effects, among them the magnetic birefringence of gases and liquids. Work on such measurements is in progress. A publication describing this device is in preparation.

3. The Anisotropy of the Molecular Optical Activity (R. L. Disch, D. Sverdlik)

The method of harmonic exclusion⁽²⁾ proposed for the determination of the anisotropy of molecular optical activity is being extended to the case of absorbing media. This case presents considerably more complexity than does that of the transparent medium, for the effects of linear and circular dichroism must be accounted for as well as those of linear and circular birefringence in an optically active absorbing anisotropic medium. This analysis is of interest in the study of optical rotatory dispersion and circular dichroism, for it affords the possibility of a more accurate characterization of the electronic transitions which are responsible

for the Cotton effect, especially in large or complex molecules, such as polypeptides.

*This research was also supported by the Army Research Office under Contract DA-31-124-ARO-D-305.

(1) M. F. Vuks, Optics and Spectroscopy 20, 361 (1966).

(2) CRL Progress Report, April 30, 1966, p. 44.

G. LASER SPECTROSCOPY OF FLUIDS*

(D. Balzarini, R. Bulos, L. Seigel, L. Wilcox)

A striking interference pattern is observed when a laser beam is passed through a fluid near its critical point. A photoelectric trace of this F-plane pattern is shown in Fig. 21. A qualitative explanation for the phenomenon illustrated in Fig. 21 may be given with the help of Fig. 22. Near the critical point, the coefficient of isothermal compressibility attains a large value; so large, in fact, that the fluid is appreciably compressed by its own weight. The density distribution which results in the earth's field is represented in Fig. 22. The fluid refracts an incident plane wave downward. Above and below the region of maximum refraction (0) are regions which refract the beam through smaller but identical angles. Interference occurs between two such ray bundles labeled (+) and (-) which combine in the focal plane of the lens L_1 . It is the systematic change of optical path difference with angle that produces the F-plane pattern illustrated in Fig. 21. No allusion to this interference pattern has been found in the existing literature.

The explanation just given permits derivation of quantitative relationships between the location of the s^{th}

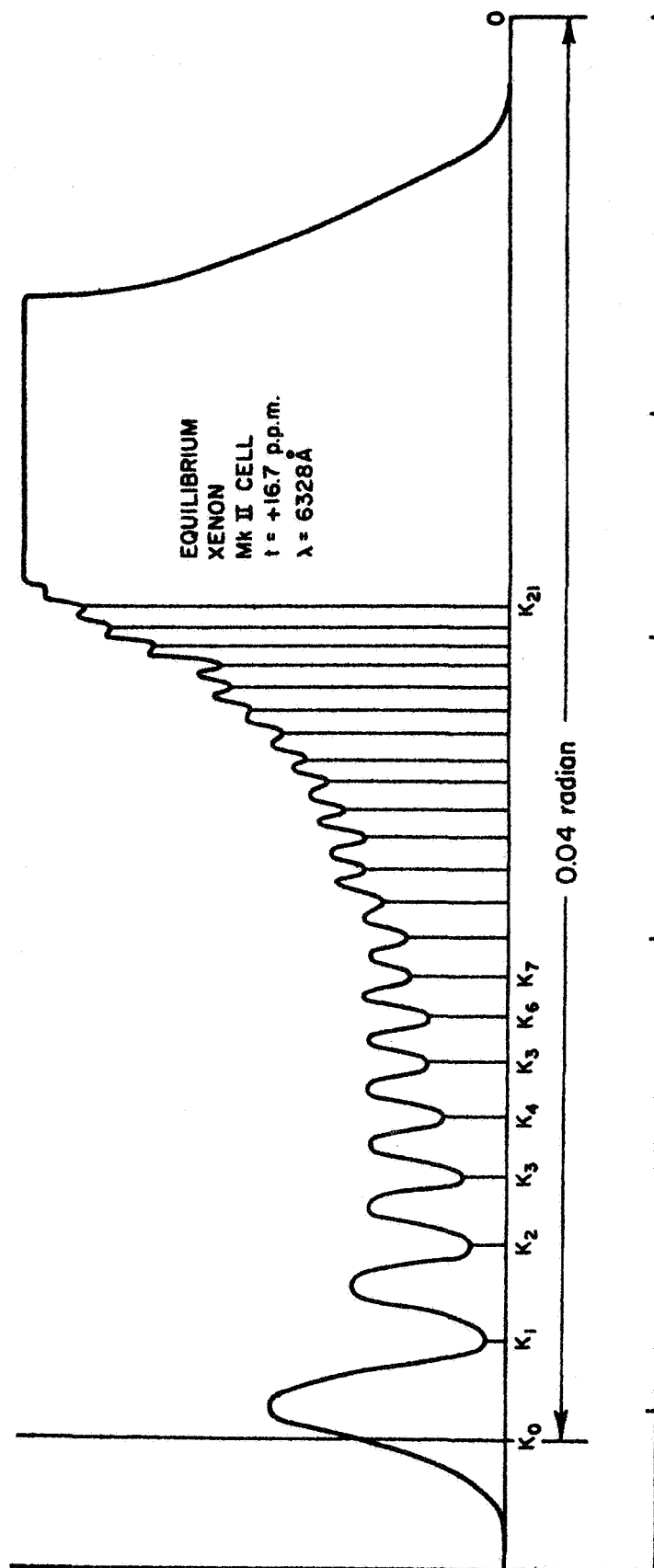


Fig. 21 Photoelectric scan of F-plane interference pattern from xenon.

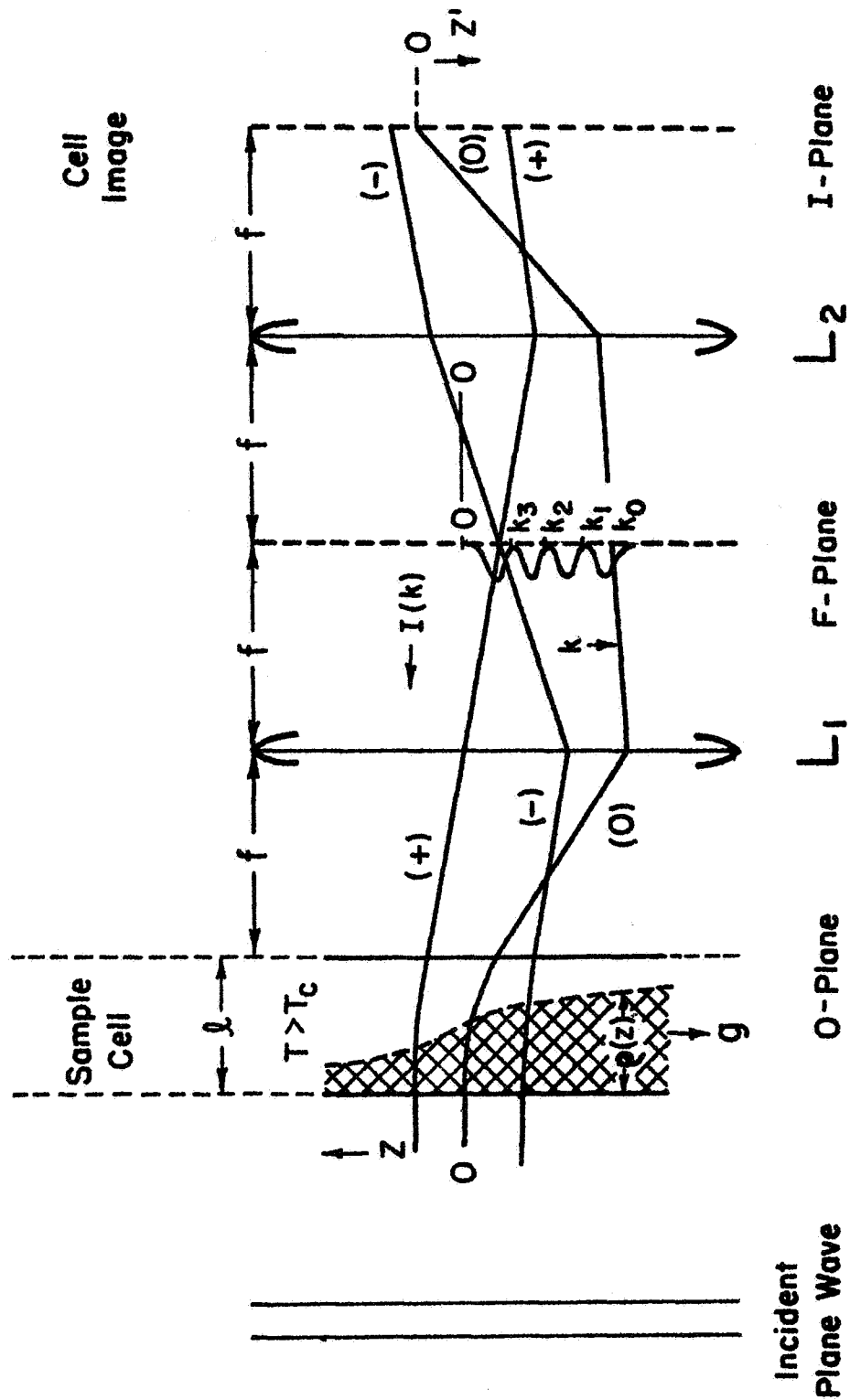


Fig. 22 Ray diagram illustrates the formation of an interference pattern by refraction through a compressible fluid in the earth's field.

minima K_S and the density as a function of height. A remarkable amount of important information can be derived from a detailed study of the interference pattern. Isotherms above and below the critical temperature can be deduced, and the shape of the liquid-gas coexistence boundary can be determined.

To illustrate the kind of results obtainable, we present Fig. 23, which shows a xenon isotherm on a pressure-density plot. The isotherm appears as the envelope of a family of experimentally determined lines — one for each minimum K_S . The isotherm is found to fit a function of the form

$$(P/P_O) = (\rho/\rho_O)^4 + (\rho/\rho_O) \quad . \quad (1)$$

Scale factors P_O and ρ_O are determined from the fit. An alternative way of treating experimental data is to plot the function $S^{3/4}K_S^{1/4}$ vs K_S . If the exponent in Eq.(1) is truly 4, the result should be a straight line. Such a plot is shown in Fig. 24 for various temperatures. Scale factors P_O and ρ_O are determined from the intercepts on such a plot. Tabulation of results for various temperatures show how P_O and ρ_O vary with temperature. Completion of the results should furnish information of increased precision bearing upon the equation of state for xenon in the immediate neighborhood of its critical point.

*This research was also supported by the Army Research Office under Contract DA-31-124-ARO-D-296.

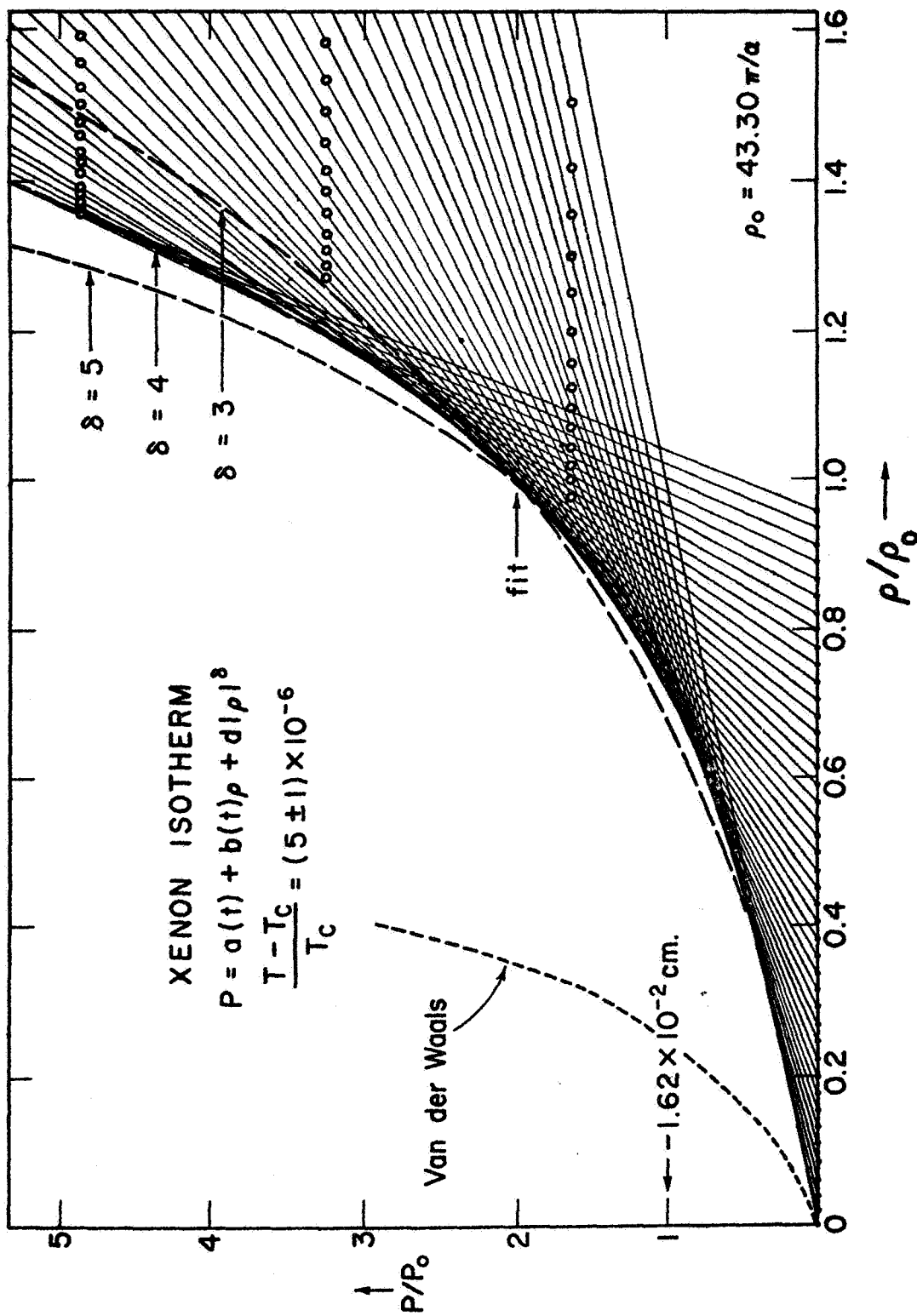


Fig. 23 The density in xenon as a function of height is the envelope of a family of lines determined by measurements of minima, K_S , in the F-plane pattern.

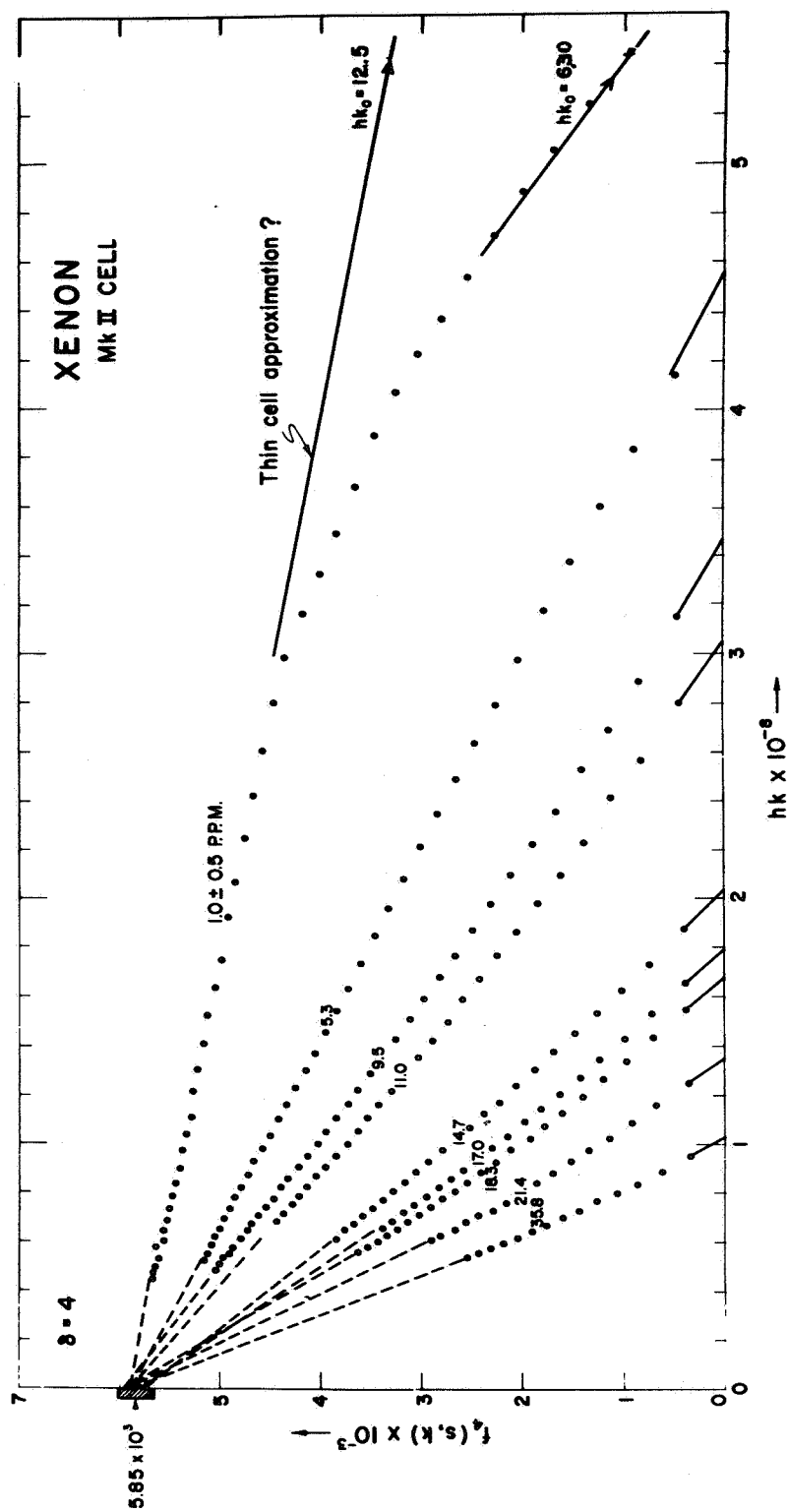


Fig. 24 Xenon isotherms. If the exponent in Eq. (1) were exactly 4, the plots for various temperatures should be linear with a slope proportional to the temperature difference ($T_C - T$).

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III. 3 SOLID STATE PHYSICS

A. INTERACTION BETWEEN A NEUTRAL BEAM AND A CONDUCTING SURFACE*

(P. Kusch, D. Raskin)

In this experiment we measure the deflection of a neutral molecular beam by a conducting surface. The deflection arises from the attractive interaction between the permanent electric dipole moment of the molecule and an image dipole induced in the surface. Since the force of interaction is weak, the intensities and deflection angles will be small; thus, even at a vacuum chamber pressure as low as 1×10^{-8} Torr, the background due to scattering of the beam by the residual gas in the chamber will be considerable.

The procedure for eliminating the residual gas scattering is to set the detector at a given deflection angle and measure intensity as a function of the chamber pressure from the base pressure of the chamber ($\approx 1 \times 10^{-8}$ Torr) to some higher pressure ($\approx 5 \times 10^{-8}$ Torr). The pressure is increased by admitting gas into the chamber through a very fine variable leak valve. The previous analysis of the residual gas scattering did not take into account the fact that the gas admitted into the chamber is not of the same composition as the residual gas.

The number of beam particles per second scattered into an element of solid angle $d\Omega$ is

$$dn_s = \int_{V_B} \left(\frac{d\sigma}{d\Omega} \right) n_B n_G v_r d\Omega dV, \quad (1)$$

where $d\sigma/d\Omega$ is the differential collision cross section for the beam on the residual gas,

n_B = density of the beam (particles/cm³)

n_G = density of the residual gas,

v_r = relative velocity between the beam particle and the gas particle,

dV = element of volume of the beam, and

V_B = volume containing all of the (divergent) beam except those portions that do not have a direct line of sight to the detector (because that line is blocked by the surface or another obstacle).

The number per second scattered into an area element dA_d of the detector is $dN_s = dn_s[dA_d/(r^2 d\Omega)]$, where r is the distance between dV and the detector as shown in Fig. 25.

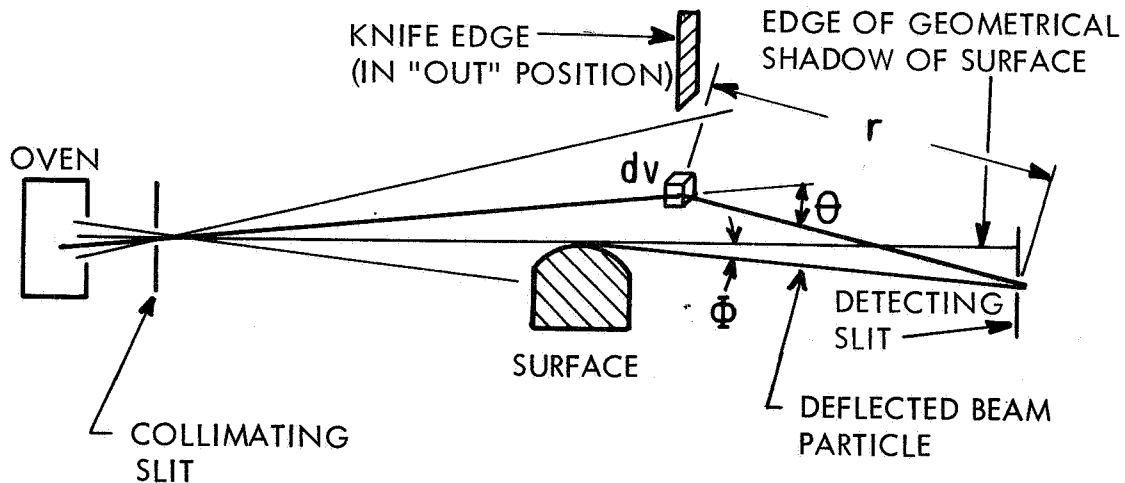


Fig. 25 Experimental layout (top view) and definition of dV and r .

Thus

$$N_s = \int_{A_d} dN_s = n_G n_B v_r \int_{A_d} dA_d \int_{V_B} \left(\frac{d\sigma}{d\Omega} \right) \frac{dV}{r^2} .$$

The number of particles per second reaching the detector when it is set at zero deflection angle is given by $N_0 = n_B \bar{v} A_d$, where $\bar{v}$ is the average velocity of the particles in the beam. Also, n_G is given by $n_G = P/kT$, where P = residual gas pressure, k = Boltzmann's constant, and T = absolute temperature in the vacuum chamber.

Thus, the scattered relative intensity is

$$\frac{N_s(\Phi)}{N_0} = \frac{P}{kT} \frac{v_r}{\bar{v}} \int_{A_d} \frac{dA_d}{A_d} \int_{V_B} \left(\frac{d\sigma}{d\Omega} \right) \frac{dV}{r^2} = Pf(\Phi) ;$$

Φ = detector deflection angle (see Fig. 25). The function f depends on the differential scattering cross section, but to a rough approximation, f is proportional to the total cross section σ .

If the chamber contains two noninteracting gases, each gas scatters the beam independently of the other; thus their effects are simply additive.

$$\frac{N_s(\Phi)}{N_0} = P_1 f_1(\Phi) + P_2 f_2(\Phi) ,$$

where P_1 and P_2 are the partial pressures of gas 1 and gas 2, respectively.

In order to interpret the data, one must also account for the different sensitivities of the ionization pressure gauge for different gases. In general, $p = SP$, where p is

the pressure reading on the ion gauge, S is the sensitivity of the gauge for the gas (S is proportional to the electron ionization cross section of the molecules in the gas), and P is the actual pressure (which we cannot measure).

Now consider what happens when a chamber with a certain residual gas (gas "1") at a "gauge" pressure p_1 has a second gas admitted until the gauge shows a new pressure p . We assume that the partial pressure of gas "1" is unchanged by admission of gas "2".

$$p = p_1 + p_2 = p_1 + S_2(P - P_1) = p_1 + S_2P - \frac{S_2}{S_1} p_1 .$$

Solving for P :

$$P = \frac{p}{S_2} + \left(\frac{1}{S_1} - \frac{1}{S_2} \right) p_1 .$$

But $P = P_1 + P_2$ and we know that $P_1 = \frac{1}{S_1} p_1$. Therefore,

$$P_2 = P - P_1 = \frac{1}{S_2} (p - p_1) .$$

Thus, we can now write the scattered relative intensity in terms of pressures as read on the ion gauge,

$$\begin{aligned} \frac{N_s(\Phi)}{N_o} &= \frac{f_1(\Phi)}{S_1} p_1 + \frac{f_2(\Phi)}{S_2} (p - p_1) \\ &= \frac{f_2(\Phi)}{S_2} p + \left[\frac{f_1(\Phi)}{S_1} - \frac{f_2(\Phi)}{S_2} \right] p_1 . \end{aligned}$$

The total relative intensity measured, including the effect of the surface, is

$$\frac{N}{N_o} = \frac{N_{\text{effect}}}{N_o} + \frac{f_2}{S_2} p + \left[\frac{f_1}{S_1} - \frac{f_2}{S_2} \right] p_1 .$$

The measured intensity at a given deflection angle is plotted as a function of p . These data points lie on a straight line in accordance with the analysis. The measured intensity is equal to the interaction intensity (with no contribution due to scattering) at a point where

$$\frac{f_2}{s_2} p + \left[\frac{f_1}{s_1} - \frac{f_2}{s_2} \right] p_1 = 0$$

or

$$p \equiv p_0 = \frac{s_2}{f_2} \left[\frac{f_2}{s_2} - \frac{f_1}{s_1} \right] p_1 \quad (2)$$

Thus, if the lines of measured intensity versus p are extrapolated to $p = p_0$, the scattering is eliminated. The only problem then is to determine p_0 . There are two ways in which to do this:

1) For a surface with large radius ($r_0 \geq 10$ cm), the intensity due to the interaction between the surface and the beam drops sharply to zero at an angle of approximately 4 min of arc. If the detector is placed at a larger angle, the measured intensity will be due only to scattering so that the extrapolation line will cross the p axis at $p = p_0$. This crossing point is independent of angle for any angle larger than the cutoff angle.

2) A knife-edge parallel to the axis of the surface is brought into the beam on the side opposite to the surface (see Fig. 25). It is placed at a point such that the direct intensity at the edge of the surface (N_0) is undiminished. This knife-edge has no effect on the deflected beam except to remove some of the scattering. The effect of the presence of

the knife-edge is to reduce the volume of integration V_B in Eq.(1). If data are taken with the knife-edge "in" and these data are plotted in the same manner as the data with the knife-edge "out," then the two lines so formed will cross at $p = p_0$, $N/N_0 = N_{\text{effect}}/N_0$. This is because the two lines differ only in the amount of scattering detected; when the scattering is eliminated (at $p = p_0$), the two intensities will be the same. Since variations in p_0 can be measured as the angle ϕ is changed, this method is the better of the two.

The two experimental checks are in complete agreement. Furthermore, the variation of p_0 with p_1 is exactly as predicted by Eq.(2). Rough calculations of p_0 for the inlet gases A, H_2 , N_2 , and He are consistent with experimental values. Beam profiles derived from extrapolations with different inlet gases show a complete independence of the type of inlet gas.

The data that have been analyzed by the foregoing procedure show a certain amount of agreement with the theory of the interaction being studied. The intensity for large-radius cylinders cuts off at a much smaller angle than the intensity for a small radius. Also, the intensities measured are of the same order of magnitude as the theoretical predictions. The data always indicate larger radii for the cylinders than are reasonable, and it has not as yet been possible to find a mechanism that would give this kind of result although work in this direction is continuing.

Program for the next interval: More data will be taken in an attempt to find an explanation for the remaining discrepancy between theory and experiment. In addition,

further calculations will be made to investigate the influence of possible extraneous effects on the data, such as irregularities on the surface, edge effects of the cylinder, and velocity distribution within the beam.

*This research was also supported by the Army Research Office (Durham) under Grants DA-ARO(D)-31-124-G568 and DA-ARO-D-31-124-G870.

B. ADIABATIC DEMAGNETIZATION IN THE ROTATING FRAME* (H. Einbinder, S. R. Hartmann)

Quasi-equilibrium states of the F^{19} nuclear spin system in CaF_2 have been produced by performing an adiabatic demagnetization in the rotating frame followed by an irradiation with a resonant oscillating field. The magnitude of the irradiating field was greater than the system's 2-G local field due to neighboring nuclei. The spin-spin interaction energy of the system was measured after various intervals of irradiation. The value of the energy underwent transient oscillations for an interval about five times that of the 30- μ sec spin-spin relaxation time. It then settled at a value that was stable for times of this scale, but different from the value at complete thermal equilibrium in the rotating frame. When the irradiation was extended to 100 msec, a relaxation of the energy value of this quasi-equilibrium state toward the value for complete thermal equilibrium was observed.

Measurements were made for several strengths of irradiation ranging from 2.5 to 6 G. The relaxation times τ , i.e., the times required for the difference between the

quasi-equilibrium and full thermal equilibrium spin-spin energy to drop to $1/e$ of its original value ranged from 6 to 65 msec.

The data are being analyzed to determine whether the spin-irradiation interaction exchanges energy with all or just a part of the spin-spin dipolar interaction. In particular, a check is being made of the possibility that the exchange of energy involves only that part of the spin-spin interaction whose Hamiltonian commutes with the spin-irradiation Hamiltonian. In this case the remaining non-commuting part could act as a perturbation facilitating the energy exchange without itself exchanging energy.

With suitable assumptions, a theoretical expression for the variation of τ with irradiating field strength of the form

$$\tau \propto \frac{h_L^2}{H^2 + h_L^2} \exp(H^2/A)$$

may be obtained. H is the effective magnitude of the irradiating field, $h_L = \text{Tr } \mathcal{K}^2 / \gamma^2 \hbar^2 \text{Tr } I_z^2$, where $\mathcal{K}$ is the Hamiltonian of the portion of the spin-spin interaction exchanging energy with the spin-irradiation interaction, γ is the gyromagnetic ratio, and A is a constant which is dependent on the spin system and is approximately equal to h_L^2 .

By determining whether the value of h_L corresponding to the entire spin-spin Hamiltonian or the value corresponding to just the part that commutes with the spin-irradiation Hamiltonian fits the data best, we might be able to determine

what division of the spin system into thermodynamic systems characterized by a temperature is valid for the case of strong irradiation.

The present data approximately fit the above form with h_L^2 of the order of $4G^2$ and $A = 4G^2$. Error caused by spin-lattice interaction prevented an exact determination of how the spin system should be divided into thermodynamic systems.

Program for the next interval: Further measurements will be made on purer specimens in an attempt to reduce the effect of spin-lattice interaction. This should permit a more accurate determination of how spin-spin energy is exchanged with the energy of magnetization along the irradiating field as a spin system approaches thermal equilibrium while subjected to strong irradiation.

*This research was also supported by the National Science Foundation under Grants NSF-GP 3379 and NSF-GP 6312.

C. NUCLEAR MAGNETIC RESONANCE IN FINE PARTICLES*

(S. R. Hartmann, G. R. Mather)

The superconducting magnet has been installed and tested, and the electronic gear necessary to utilize its high (≈ 50 kG) field is nearly complete. Lithium and platinum require resonance frequencies between 40 and 80 Mc/sec at a field of 50 kG, and since the present equipment operates to only 20 Mc/sec, a transmitter and receiver capable of operating at the higher frequencies are needed.

The high field available from this magnet has the obvious advantage of increasing the signal-to-noise ratio from the sample, an advantage which is important for this

work because of the small amount of sample available. It may also prove to be important to the extent that fine-particle effects depend on the applied field, since it will allow this parameter to be varied over a range much wider than with a conventional magnet.

The work to date with this magnet has been done at a field of 23 kG, corresponding to a platinum resonance frequency of 21 Mc/sec which is the upper limit of our present transmitter. The sample consists of the 100-Å-size platinum black which has been heated to $\approx 500^\circ\text{C}$ under a helium atmosphere and then allowed to cool slowly. After this treatment, the resonance signal is observable at the field expected for bulk platinum, with a signal-to-noise ratio which agrees well with the calculated value.

While it is possible that the heating has caused sintering of the platinum particles, preliminary data at temperatures of 4.2°K and below indicate a possible anomaly in spin-lattice relaxation, to the extent that the decay is not strictly exponential and that the decay rates are generally too long. It is still too early to draw any conclusions, however, and we are currently working to discover and correct any defects in equipment or technique which might give rise to anomalous results. In addition, we plan to study the sample under an electron microscope to determine the extent to which sintering has occurred.

As mentioned previously,⁽¹⁾ we have been able to produce lithium particles of less than 1μ in size by ultrasonic dispersion of the molten metal. There results, however, a distribution of particle sizes, and the chief problem has

been to separate out particles of the desired size. The availability of filters with pore openings of $100 \text{ \AA} \pm 40\%$ should make this problem solvable, and it should prove possible to produce a good small-particle lithium sample with reasonably uniform particle size.

Program for the next interval: Work with the treated platinum black will continue, and data on spin-lattice relaxation will be taken at various temperatures and field strengths.

*This research was also supported by the National Science Foundation under Grants NSF-GP 3379 and NSF-GP 6312.

(1) CRL Progress Report, April 30, 1966, p. 71.

D. NUCLEAR DOUBLE RESONANCE IN KClO_3 *

(S. R. Hartmann, E. P. Jones)

An extensive search for the K^{40} signal was made in a KClO_3 sample with natural abundance K^{40} (0.012%) and with enriched K^{40} (2.8%). Several double-resonance signals, mostly unexplained but not due to K^{40} , were observed. A complete interpretation has not yet been made of the results of these experiments.

*This research was also supported by the National Science Foundation under Grants NSF-GP 3379 and NSF-GP 6312.

E. PHOTON-ECHO RESONANCE *

(S. R. Hartmann, N. A. Kurnit)

Initial observations of photon echoes in ruby indicated the necessity of applying a magnetic field along the optic axis of the sample to inhibit a relaxation process. Recent investigations of photon echoes have been directed toward understanding the mechanism responsible for this relaxation.

It has now been possible to reduce the saturation effect of closely spaced excitation pulses sufficiently to enable the observation of photon echoes in zero applied magnetic field. Also, by utilizing a sample whose optic axis is oriented perpendicular to the direction of propagation of the light, it has been possible to place the sample in an electromagnet capable of producing a field of several kilogauss. In Fig. 26 are shown the data obtained for 34 and 75 nsec pulse separations for values of the magnetic field ranging between 0 and 5000 G along the optic axis. The experimental data are well approximated by curves of the form

$$S = S_0 e^{-\alpha(\tau) [H_L^2 / (H_L^2 + H_Z^2)]},$$

where α is a parameter depending only on the pulse separation τ , and the field $H_L \approx 15$ G is comparable to the ESR linewidth. Superimposed upon these curves are two sharp, pronounced dips in echo amplitude at 2 and 4 kG associated with level crossings in the ground state.⁽¹⁾

Earlier experiments have shown that as the magnetic field is tilted away from the optic axis, the echo amplitude falls off nearly exponentially with angle. When the magnetic field is increased to above 1 kG, it is now found that a transverse component of the magnetic field is no longer as effective in destroying the echo. This is illustrated by the 220-nsec data in Fig. 26 in which the magnetic field is misaligned by 2° from the optic axis. The rise in echo amplitude at ≈ 2 kG is believed to be associated with the applied magnetic field at the Al sites becoming larger than the effective field due to the Cr^{3+} ions at these sites.

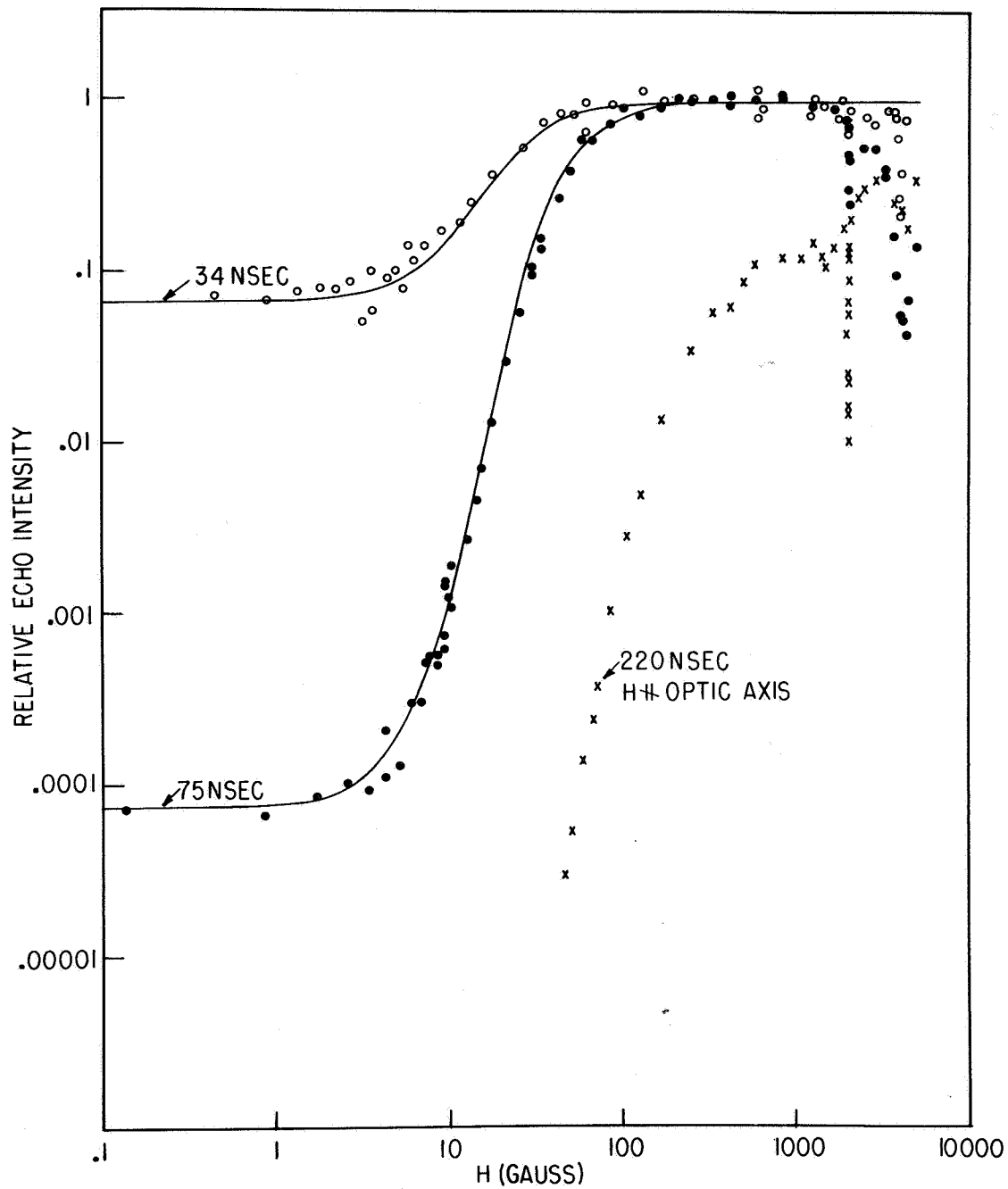


Fig. 26 Amplitude of photon echo as a function of applied magnetic field. Field is parallel to optic axis for 34 and 75 nsec pulse separations and is misaligned by 2° from optic axis for 220 nsec separations.

A proper treatment of the photon-echo relaxation involves a density matrix calculation that takes into account the multiplicity of the levels associated with the ground and excited states. A general calculation that neglects interaction of Al nuclei shows that the application of a magnetic field of any magnitude and orientation does not relax the echo. This problem is solvable in closed form and shows that after a small transient effect which will average out if the field is inhomogeneous, one always obtains an unrelaxed echo. The inclusion of Al interactions to provide time-dependent relaxation fields at the Cr^{3+} sites complicates the problem, but two important features can be displayed. When the applied magnetic field becomes larger than the internal local fields at the Cr^{3+} sites, one must eliminate interactions in which the Cr^{3+} electron flips in the applied field as this does not conserve energy. When the applied field is larger than the hyperfine fields at the Al nuclei, those interactions involving Al spin flips which do not conserve energy in this field must also be eliminated. The successive elimination of certain spin-spin interactions at higher fields reduces the effectiveness of the echo dephasing mechanism, and consequently, one observes larger echo amplitudes as the magnetic field is increased, particularly at fields corresponding to: 1) the local field at the Cr^{3+} sites and 2) the local field at the Al sites.

Preliminary results showing the relaxation of the stimulated echo are given in Fig. 27. These results exhibit the expected relative independence of the echo amplitude on the separation of the last two excitation pulses.

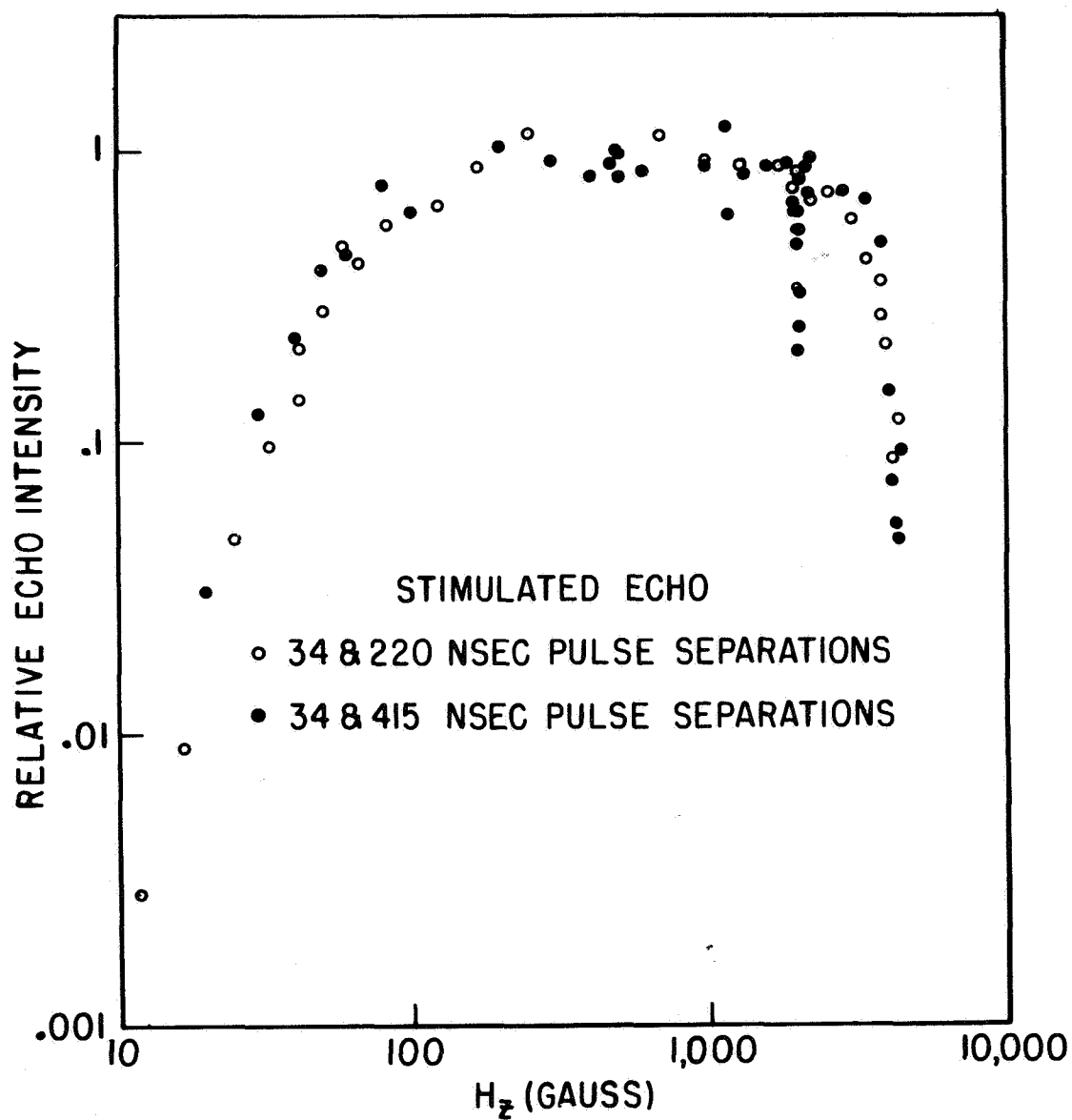


Fig. 27 Amplitude of stimulated echo as a function of magnetic field applied along optic axis, first two pulses separated by 34 nsec, first and third by 220 and 415 nsec.

We have also observed photon echoes polarized parallel to the optic axis of the ruby by utilizing excitation pulses polarized in that direction. These new echoes are two orders of magnitude smaller than the regular echoes, due in part to their smaller dipole matrix element, and are enhanced by the application of a small magnetic field along the optic axis in essentially the same way as the echoes produced in the standard way. An important feature of these results was that the amplitude of the new "parallel" echoes as a function of the angle between the magnetic field and the optic axis was essentially the same as that observed with the ordinary perpendicular echoes.

Program for the next interval: The above measurements will be extended to a wider range of pulse separations.

*This research was also supported by the Army Research Office under Contract DA-31-124-ARO-D-341.

(1) E. O. Schulz-Dubois, Bell System Tech. J. 38, 271 (1959).

F. BRILLOUIN SCATTERING*

(D. Grischkowsky, S. R. Hartmann)

Work is continuing on the improvement of the experimental arrangement for the investigation of the phonon bottleneck effect described previously.⁽¹⁾ Effort is presently being directed toward 1) modifying the ruby laser to operate in the Q-switched mode, 2) finding a suitable crystal sample, and 3) reducing extraneous scattering of the laser beam.

When the experimental setup is completed, preliminary attempts will be made to heat the paramagnetic spin system by inverting the spin population, and to detect the phonon avalanche. (2)

Program for the next interval: If the preliminary work is successful, an effort will be made to observe the resonant phonons by monitoring the Brillouin scattering of the laser beam by the paramagnetic crystal.

*This research was also supported by the Army Research Office under Contract DA-31-124-ARO-D-341.

(1) CRL Progress Report, December 15, 1965, p. 52.

(2) W. J. Brya and P. E. Wagner, Phys. Rev. Letters 14, 431 (1965).

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IV.3 RADIO AND X-RAY ASTRONOMY 8/10/10
N67-26620

A. MODELS OF PLANETARY ATMOSPHERES*
(W. Ho, I. Kaufman, P. Thaddeus†)

1. The Atmosphere of Venus

The microwave absorption coefficients measured for mixtures of carbon dioxide with nitrogen and argon⁽¹⁾ and water vapor with nitrogen⁽²⁾ have been applied to an analysis of the observed microwave emission spectrum of Venus with regard to the composition and structure of its atmosphere.

Microwave emission from Venus has been reported over the frequency range from 0.4 Gc/sec to 300 Gc/sec. At frequencies below 10 Gc/sec the intensity of emission is consistent with that expected from a blackbody at approximately 600°K; between 10 and 100 Gc/sec the brightness temperature decreases to about 300°K, and varies only slightly from this latter value as the frequency is increased further. Observations of limb darkening and the distribution of polarization over the planetary disc, together with the independence of brightness temperature with time and, at low frequencies, with frequency, lead to the conclusion that the centimetric radiation originates at the surface of the planet and is thermal in origin. The transition in brightness temperature in the vicinity of one-centimeter wavelengths indicates some atmospheric constituent whose opacity increases appreciably with frequency in this region.

From spectroscopic evidence, the only gases definitely identified in the Venus atmosphere are carbon dioxide and, in very small amounts, water vapor. The larger part of the

atmosphere is made up of spectroscopically undetectable gases, which, from astrophysical considerations, are most likely nitrogen, argon, and neon. The study of pressure-induced microwave absorption in mixtures of these gases was the primary aim of this work.

The model of the Venus atmosphere used in our calculations consists of a lower layer having an adiabatic lapse rate from the surface to the cloud tops, and an isothermal layer above the cloud tops. Within each layer the temperature and pressure are then known functions of altitude, surface pressure, surface temperature, cloud-top temperature, scale height, and specific heat ratio of the atmospheric gases. From the radio observations and radar determinations of surface emissivity, we have assumed the surface temperature to be 675°K; from infrared observations, we have taken the cloud-top temperature to be 250°K.

Our results of the laboratory measurements of microwave absorption coefficients can be combined into a single approximate expression accurate to within a few percent:

$$\alpha = P^2 \bar{\nu}^2 (273/T)^5 (15.7 f_{\text{CO}_2}^2 + 3.90 f_{\text{CO}_2} f_{\text{N}_2} + 2.64 f_{\text{CO}_2} f_{\text{A}} + 0.085 f_{\text{N}_2}^2 + 1330 f_{\text{H}_2\text{O}}) \times 10^{-8}, \quad (1)$$

where α = absorption coefficient, in cm^{-1} ,

P = pressure in atmospheres,

$\bar{\nu}$ = wave number,

T = temperature in °K,

and f_{CO_2} , f_{N_2} , f_{A} , $f_{\text{H}_2\text{O}}$ = molar fractions of CO_2 , N_2 , A, and H_2O , respectively.

The term due to quasi-resonant absorption in water vapor was measured in nitrogen, where N_2 - H_2O collisions primarily determine the linewidth, and this term strictly applies only to atmospheres where nitrogen is the main constituent. At pressures lower than a few atmospheres, the 1.35-cm water-vapor line makes a resonant contribution to the absorption not included in Eq.(1).

The average brightness temperature of the planet as a function of frequency was calculated, and the best fit to the radio observations led to a relation between surface pressure and atmospheric composition shown graphically in Fig. 28. The lower solid line corresponds to a dry CO_2 - N_2 atmosphere, and the upper solid line to a dry CO_2 -Ne atmosphere. For an arbitrary mixture of N_2 , Ne, A, or He, as the unobserved major fraction of the atmosphere, the theoretical curve lies between the two limits. Also included in Fig. 28 is the ground pressure as a function of the fraction of water vapor if pressure-broadened resonant absorption of water vapor in N_2 is the source of the Venus microwave opacity.

Our conclusions drawn from these results are:

- 1) If induced absorption in a CO_2 - N_2 atmosphere, where CO_2 is a relatively minor constituent, is the source of the Venus microwave opacity, and if the lapse rate is close to adiabatic, then the surface pressure lies in the range from 100 to 300 atmospheres.

- 2) Water vapor can account for the microwave opacity of the atmosphere only if water is more abundant by several orders of magnitude than the infrared spectrographic studies suggest.

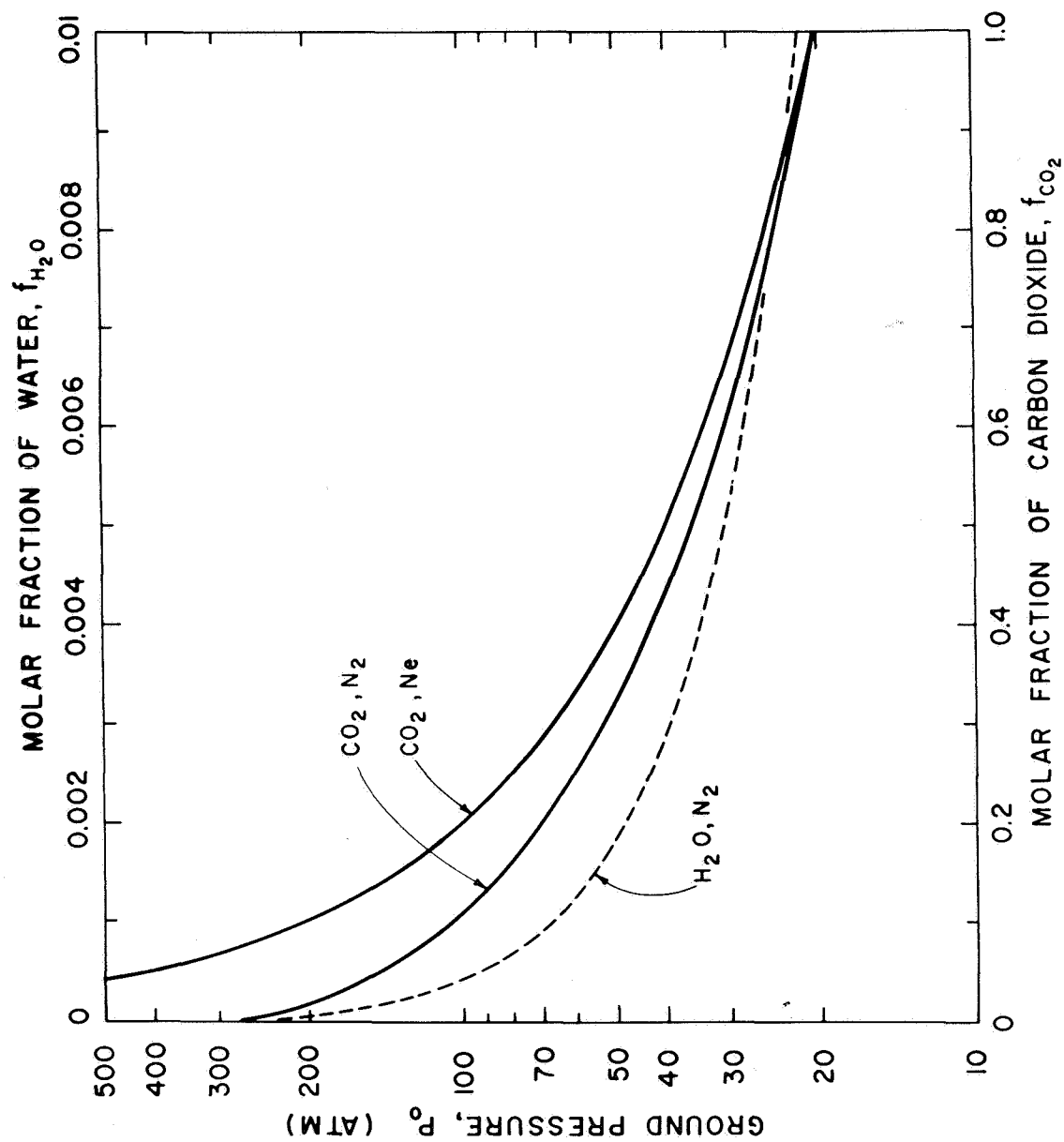


Fig. 28 Surface pressure of Venus as a function of atmospheric composition.

2. Microwave Absorption in Oxygen

Current attempts to measure the cosmic background radiation at low microwave frequencies with ground-based equipment require knowledge of the emission from the earth's atmosphere. Nonresonant emission by molecular oxygen is the main source of the low-frequency emission, and is comparable in magnitude to the cosmic radiation at 20.7 cm.⁽³⁾ Accurate determination of the absorption in the low-frequency wing of the oxygen spectrum over a wide range of temperatures is required to properly interpret these experiments.

Practically all experimental measurements of microwave absorption by oxygen have been made in the region around 60 Gc/sec, where oxygen exhibits resonant absorption. At these frequencies measured values of linewidths at low pressures, where individual lines are resolved, disagree with linewidths necessary to explain the absorption at one atmosphere. Similar disagreements have been observed at low frequencies, where the absorption is largely nonresonant. In order to obtain a greater body of experimental data with which to compare the various theoretical treatments, we are currently measuring the absorption due to pure oxygen, air, and an oxygen-argon mixture at 9260 Mc/sec.

The loss tangent of pure oxygen was measured for pressures up to 17 atm at temperatures over the range from 60°C to -78°C. At all of these temperatures, ϵ''/ρ increases almost linearly with density up to 6-8 Amagat, then tends to level off; this behavior is roughly consistent with the variation expected from a nonresonant line shape in this pressure range. The results were compared with the

theoretical values calculated from the equations of Van Vleck⁽⁴⁾ and Gordon,⁽⁵⁾ and the value of linewidth which gave the best fit to the experimental points was determined for each case.

The Van Vleck expression described the absorption reasonably well up to a density of about 10 Amagat at each temperature, but predicted absorptions much less than those observed at higher densities; the Gordon formula gave reasonable agreement up to the highest pressures measured except at the very lowest temperatures, where the higher density absorption was somewhat higher than predicted.

The half-widths at each temperature which result in the best fit of the Van Vleck expression to the low density points are 20 to 25 percent higher than the half-widths giving the best fit of the Gordon expression, although the temperature variation of the two half-widths appears to be the same.

Figure 29 shows the variation of ϵ''/ρ with ρ for pure oxygen at 25°C and the "best fit" curves predicted by the two theories. The half-widths used to compute the curves are 811 Mc/sec/atm for the Van Vleck curve and 684 Mc/sec/atm for the Gordon curve.

Work will continue on this phase of the program, extending the temperature range, and using mixtures of oxygen with nitrogen and argon.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-442.

†NASA Institute for Space Studies.

(1) CRL Progress Report, June 15, 1965, p. 58.

(2) CRL Progress Report, April 30, 1966, p. 78.

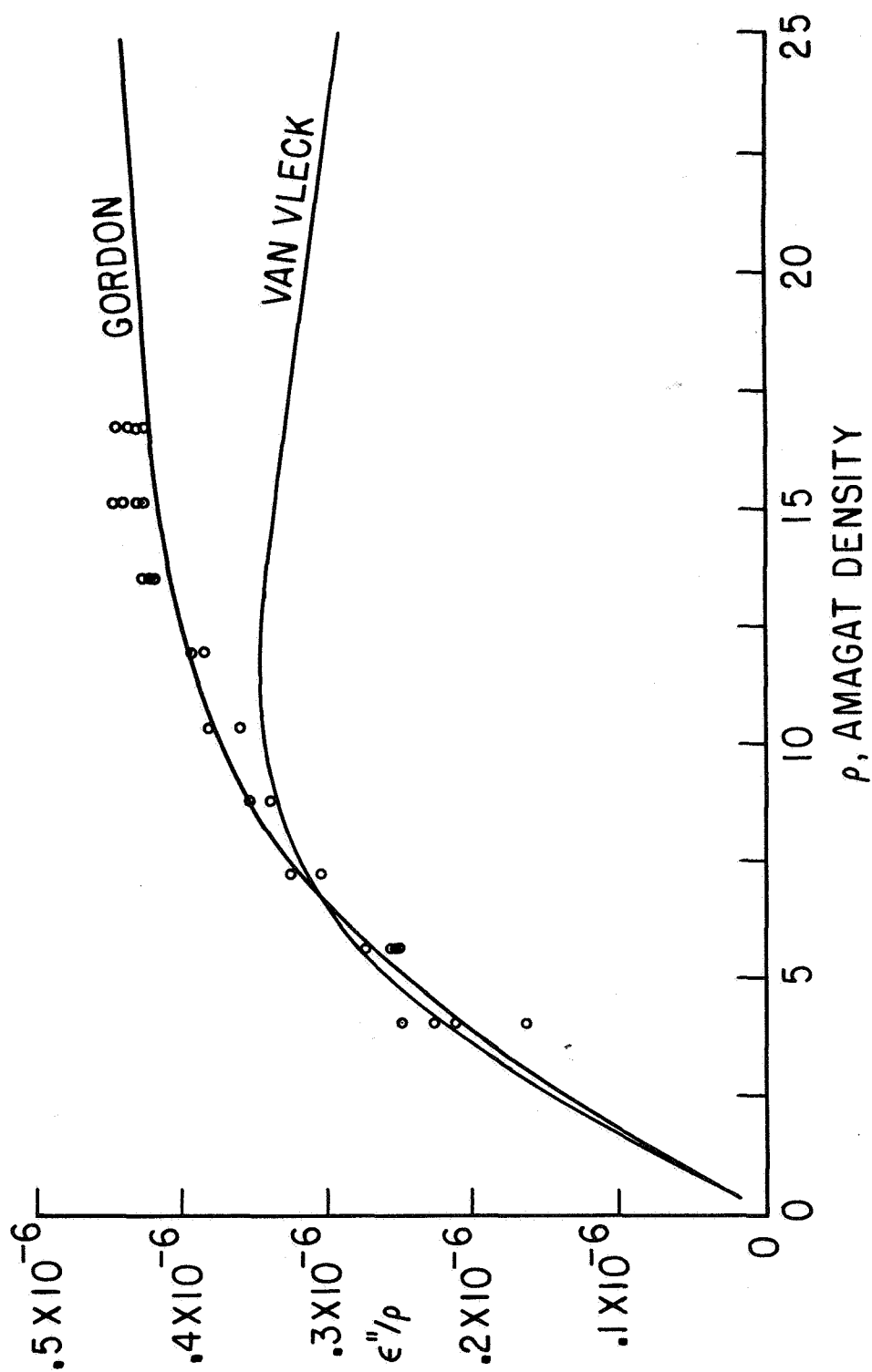


Fig. 29 Absorption by oxygen at 9260 Mc/sec, 25°C.

(3) T. F. Howell and J. R. Shakeshaft, Nature 210, 1318 (1966).

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(5) R. Gordon, J. Chem. Phys, to be published.

B. X-RAY POLARIZATION*

(T. Wing, F. Kantor, R. Novick)

The development of proportional counters continued during this period. The first requirement on the counters is the ability to withstand the space environment. A second requirement is associated with the pointing accuracy of the guidance system. The experiment is performed by rotating the apparatus while pointing it directly at the x-ray source. The guidance system has an accuracy of only a few degrees. It is necessary that this lack of accuracy does not result in a false polarization indication.

To satisfy the first requirement, the counters were successfully redesigned to withstand vacuum and temperature cycling. Because of the three-atmosphere pressure of xenon within the counters, this proved to be quite difficult.

The second requirement was fulfilled by redesigning the counters to maintain constant efficiency of the apparatus as the angle between the incident x-ray beams and the axis of the apparatus is changed from zero to a few degrees.

Program for the next interval: A balloon flight is planned in order to measure the background counting rate and the background reduction obtainable by use of a plastic anti-coincidence shield and by rejection of events in which more than one counter fires.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-445, Scope B.

C. X-RAY TELESCOPE*

(F. Kantor, R. Novick, T. Wing)

X-Ray Focusing Systems

A large-area x-ray collector has been partially completed and is tentatively scheduled for flight in an Aerobee 150 sounding rocket next spring. The system has been checked mechanically for vibration resistance and found to withstand 30 g of random vibration at 20-2000 cps. Individual modules are able to withstand 100 g of sinusoidal vibration at 100-2000 cps, with the lowest resonance, as determined by tests at lower acceleration, well above 100 cps. The collector was not tested at higher acceleration because of limitations in the shaker system.

A computer program has been prepared to calculate the necessary machining dimensions for fabricating a collector of the type constructed. This program has been expanded, and a revised version, including a plot of the contribution of each module to the focused image, a calculation of effective area as a function of energy and angle off axis, and generation of a logarithmic contour plot of the image plane for an image off axis and out of focus, is nearly completed.

Preliminary results indicate that it will be possible to resolve the image into several regions, thereby permitting use of the collector in a system which tracks an x-ray source.

Construction of the necessary electronic equipment and detector systems is progressing; completion is expected in February.

We are presently selecting astronomical x-ray sources for observation during the first flight, with several alternatives in case of delay in launch.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-445, Scope B.

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²Asst Professor of Physics, Department of Physics and Astronomy,
University of Florida, Gainesville, Florida.

³Asst Professor of Physics, Department of Physics and Astronomy,
University of Toledo, Toledo, Ohio.

⁴Research Physicist, North American Aviation Science Center,
Thousand Oaks, California.

⁵Asst Professor of Physics, Department of Physics, Brown Univer-
sity, Providence, Rhode Island.

⁶Research Associate, Lawrence Radiation Laboratory, University
of California, Livermore, California.

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13. ABSTRACT (cont'd)

Induced magnetic dipole transitions between the Zeeman sublevels of the 2^3P_2 , $F = 7/2$ state of singly ionized lithium have been observed at 50 MHz. These and other Zeeman resonance experiments at low frequencies and low magnetic fields provide a basis for subsequent measurements of the fine-structure separations and for an eventual redetermination of the fine-structure constant α .

The Zeeman transitions in metastable autoionizing atomic states of potassium have been thoroughly studied; in particular, the excitation functions and power dependence have been determined. Some direct hyperfine transitions were observed giving a $^4F_{9/2}$, $F = 6$ to $F = 5$ splitting of 568.8 ± 0.1 MHz. The apparatus has recently been modified to increase the accuracy with which the magnetic field can be determined, and thereby to obtain an improvement of two orders of magnitude over the previously reported measurements.

Experiments have been carried out to investigate the properties of off-resonant light as a probe of optically pumped vapors. Magnetic resonance was detected in an optically pumped rubidium vapor using light from the Sr II spectrum. No depumping of the vapor or attenuation of the probing beam could be observed.

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