

## V. RADIO-FREQUENCY SPECTROSCOPY

### A. MOLECULAR-BEAM RESEARCH

Prof. J. R. Zacharias	Dr. J. Levinson
Prof. B. T. Feld	Dr. Hin Lew
Dr. D. E. Nagle	Dr. C. W. Zabel
Dr. L. Davis, Jr.	J. T. Eisinger
Dr. G. Knight	T. M. Hahn, Jr.

W. B. Pohlman

#### 1. Third Molecular-Beam Apparatus

Work is continuing in the construction of this apparatus.

Dr. Hin Lew

#### 2. Nuclear Spin and Hyperfine Structure of Cs<sup>135</sup> and Cs<sup>137</sup>

A mixture of cesium isotopes produced by uranium fission was obtained from the Oak Ridge National Laboratory. In the oven of a molecular-beam apparatus were placed .25 millicuries of this mixture to which 500 micrograms of stable Cs<sup>133</sup> had been added. A molecular-beam magnetic resonance experiment was performed, using a mass spectrometer to sort out the isotopes. Resonance transitions were observed for mass numbers 133, 135, and 137. First results indicate that the nuclear spin of 135, like that observed for 133 and 137, is  $7/2$ ; that the sign of the nuclear magnetic moment is positive; and that the hyperfine structure  $\Delta\nu = 9723.00 \pm 50$  megacycles. From Kusch, Millman and Rabi's value of  $\mu_{133} = 2.558$  nuclear magnetons, the nuclear magnetic moment of Cs<sup>135</sup> is  $(2.706) \pm .5$  per cent nuclear magnetons (without diamagnetic correction).

Dr. D. E. Nagle

#### 3. The Fourth Atomic-Beam Apparatus

Machine work on the main vacuum system has been completed; the vacuum system has been set up, and pumps installed. The location of leads in the apparatus is now in progress.

The construction of deflecting magnets is in progress, and machine work on the two mass spectrographs has been completed. The construction of amplifiers, and other electronic circuits to be used with the apparatus has been nearly completed.

T. M. Hahn, Jr.

#### 4. An Electron-Bombardment Ionizer

The problem of detection has restricted the choice of elements for study by the molecular- and atomic-beam magnetic resonance methods. For example, one would hesitate to investigate manganese using the current

(V. RADIO-FREQUENCY SPECTROSCOPY)

methods of detection, yet  ${}_{25}\text{Mn}^{55}$  would be an extremely interesting nucleus to investigate, since preliminary calculations indicate that the atom has an electronic structure which offers the possibility of detecting an octopole moment in the nucleus. Many other nuclei, too, cannot be attempted for lack of a suitable method of detection.

Therefore, a new method of detecting atomic and molecular beams is being investigated. The beam to be detected is bombarded by an electron beam moving perpendicularly to it. Calculations indicate that a large proportion of the atoms could be ionized, and then continue to move in substantially the same direction. The ions are then accelerated electrostatically for passage through a mass spectrograph, and finally counted by hitting the first plate of an electron multiplier.

A simple model of such a device has been built. In a test run it was found that excessive background exists, apparently due to ions formed when the electron beam strikes the anode. The model is now being modified so as not to admit ions formed at the anode into the mass spectrograph.

Dr. J. Levinson

B. NUCLEAR MAGNETIC RESONANCE

Prof. F. Bitter  
N. I. Adams, III

W. Dickinson  
T. Wimett

1. Nuclear Magnetic Moments and Hyperfine Structure  
of the Rubidium Isotopes

A recent determination (1) of the nuclear moments of the rubidium isotopes indicates that, as was first suspected by Millman and Kusch (2), the observed ratio of the magnetic moments does not agree with the ratio calculated from hyperfine structure,

$$\frac{\mu_{\text{Rb}^{87}}}{\mu_{\text{R}^{85}}}$$

Observed	(1)	$2.0327 \pm 0.001$
Observed	(3)	$2.038 \pm 0.01$
Calculated	(2)	$2.0261 \pm 0.0003$

The interaction between the optical electron and the nucleus is of two kinds - electrostatic and magnetic - and it is of interest to determine whether the observed effect can be accounted for on the basis of either of these two forces. The effect of spreading the nuclear charge, usually

## (V. RADIO-FREQUENCY SPECTROSCOPY)

considered concentrated at a point at the origin, over a finite sphere of radius  $R_0$ , is to reduce the electronic density  $\psi^2(0)$  at the nucleus. This effect was shown by Rosenthal and Breit (4) to lead to a correction in calculating hyperfine structure of the order of  $zR_0/a_0$  for atoms having a nuclear charge in the range of rubidium. The  $a_0$  is the radius of the first Bohr hydrogen orbit. While this ratio is 0.5 per cent for rubidium, and can account for discrepancies in the hyperfine structure of the order of the observed discrepancy (0.3 per cent), it is very unlikely that this is the explanation, as it would require an enormous difference in size of  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$ . Besides the fact that all available evidence supports a slowly varying nuclear radius proportional to the cube root of the mass number, we have specific evidence (5) that there is no observable isotope shift in the spectrum of rubidium, and therefore no such very large difference in nuclear size. Effects due to a magnetic structure inside the nucleus, first proposed by Kopfermann (6) in the days when such effects could well be dismissed as negligible, and recently applied by Bohr (7) to the proton-deuteron problem, also lead to a correction of the order of  $zR_0/a_0$ . This is a much more reasonable explanation, since there is no objection to assuming the wave function for the odd proton in rubidium to be quite different for the two isotopes. In fact, this might be expected on the basis of a vector model (8) of the nucleus, which would assign to  $\text{Rb}^{87}$  with  $I = 3/2$ ,  $l + s = 1 + 1/2$ , and to  $\text{Rb}^{85}$  with  $I = 5/2$ ,  $l - s = 3 - 1/2$ .

The fact that effects of this order of magnitude have not been observed for the gallium isotopes (9) or for the thallium isotopes (10) is not unreasonable since, in both cases, the two isotopes have the same nuclear spin and comparable nuclear moments, and it is therefore not unreasonable to assume that they also have comparable wave functions for the odd proton.

Prof. F. Bitter

### 2. Nuclear Magnetic Moments of $\text{Rb}^{87}$ and $\text{Rb}^{85}$

An apparatus for a precision measurement of the g-factor ratio  $\text{Rb}^{87}$  to  $\text{Rb}^{85}$  is being constructed. Other work is in temporary abeyance.

N. I. Adams, III

### 3. The Magnetic Moment of $\text{Be}^9$

The ratio of the frequency of the nuclear magnetic resonance of  $\text{Be}^9$  to that of the proton has been measured at 7000 gauss. The seventh harmonic of the  $\text{Be}^9$  frequency was heterodyned with the fundamental of the

(V. RADIO-FREQUENCY SPECTROSCOPY)

proton frequency in order to obtain higher precision in measuring the frequency ratio. Both resonances were obtained from a single sample consisting of an aqueous solution of beryllium fluoride. The sample container was a cylindrical section 1/2 inches long and 1/2 inches in diameter. The coil for the 4.2-megacycle- ( $\text{Be}^9$  resonant frequency) bridge was wound around the cylinder, and the coil for the 30-megacycle- (proton resonant frequency) bridge was wound at right angles over the first. This method assures that the external magnetic field seen by the two types of nuclei is the same.

A total of ten determinations of the frequency ratio was made. The following result was obtained:

$$\frac{\nu(\text{Be}^9)}{\nu(\text{H}^1)} = 0.1405187 \pm 0.000002 .$$

All of the ten values fall well within the above limits which were determined entirely by the uncertainty in locating the exact centers of the resonance curves.

W. Dickinson, T. Wimett

4. The Nuclear Resonance of  $\text{Li}^6$

A search was conducted for the nuclear magnetic resonance of  $\text{Li}^6$  in a  $\text{Li}^6$  enriched solution of  $\text{Li}_2\text{SO}_4$  obtained from the Atomic Energy Commission. The resonance was found after adding a small amount of paramagnetic ion to shorten the thermal relaxation time of the nuclei. Work is now in progress to make a determination of the resonance frequency ratio of  $\text{Li}^7$  to  $\text{Li}^6$ .

W. Dickinson

5. Deuteron-Proton Moment Ratio

A measurement of the ratio of the deuteron resonance absorption frequency to that of the proton has been made at 6900 gauss in the iron-core magnet. The sample used contained both nuclei in a mixture of two-thirds heavy water and one-third tap water with two drops of glycerine added to enhance the proton resonance. The two r-f coils were wound on the single sample as was done in the  $\text{Be}^9$  experiment. Frequencies employed were 4.51 Mc for deuterons, 29.4 Mc for protons, and the ratio was calculated from measurements of the deuteron frequency and the beat note obtained by mixing the thirteenth harmonic of the deuteron frequency with the second harmonic of the proton frequency. Two runs were made yielding thirteen determinations from which the following result was obtained:

$$\frac{\nu_d}{\nu_p} = 0.15350585 \pm 0.00000085 .$$

## (V. RADIO-FREQUENCY SPECTROSCOPY)

The thirteen determinations fall within the given limits of error.

This value is in agreement with a recent measurement by Bloch and coworkers. It does not agree, however, with two other similar measurements, one by Bitter, Alpert, Nagle and Poss (16) and one by Siegbahn and Lindstrom (18), although these latter two agree with each other within limits of error.

Work is in progress to further improve the precision of this measurement by means of Helmholtz coils constructed for the purpose and to carefully investigate the above discrepancies.

T. Wimett

### 6. Nuclear Magnetic Moment Values

The following is a list of all the nuclear magnetic moments which to date have been measured by the high precision nuclear induction and/or magnetic resonance absorption methods. In cases where the value of a moment has been determined by more than one investigator the value listed and its uncertainty have been weighted according to standard statistical methods.

The third column of the table lists the moments in units of the magnetic moment of the proton  $\mu_p$ . The nuclear magneton unit was not used for two reasons. First, in order to retain the high precision in the measured resonance frequency ratios, the value of the proton moment in nuclear magnetons would have to be known to equally high precision. At present this is not possible because of the relatively large probable errors associated with the physical constants used in conversion. Second, the nuclear magneton is essentially an artificial unit of magnetic moment, having no such physical significance as the Bohr magneton in the electronic case.

The uncertainty in each value in column 3 is given by the two subscript figures following the value. They indicate the uncertainty in the last two significant figures; thus  $0.7630_{70} = 0.7630 \pm 0.0070$ .

The fourth column gives literature references in order of decreasing precision of the results obtained.

The fifth column lists the diamagnetic correction as can best be calculated using the Hartree atom model plus a direct calculation using the electronic wave function in the case of hydrogen.  $H'$  is the small induced magnetic field seen by the nucleus in addition to the externally applied field  $H$ . In order to keep experimental values separate from theoretical calculations the diamagnetic correction has not been applied to the moments in column 3. It can easily be done by multiplying by the factor:

## (V. RADIO-FREQUENCY SPECTROSCOPY)

$$\frac{1 - (H'/H)_{\text{Proton}}}{1 - (H'/H)_{\text{Nucleus}}}$$

TABLE

Nucleus	Spin	$\mu/\mu_p$	References	H'/H (%)
$^1_0\text{N}^1$	1/2	- 0.685000 30	11,12	
$^1_1\text{H}^1$	1/2	1.00000---	13,14	0.0018
$^2_1\text{H}^2$	1	0.30701488 96	15,18,17,16,19	0.0018
$^3_1\text{H}^3$	1/2	1.066636 10	20,21	0.0018
$^3_2\text{He}^3$	1/2	0.7630 70	22	0.0048
$^7_3\text{Li}^7$	3/2	1.165857 48	18,1,23	0.0086
$^9_4\text{Be}^9$	3/2	- 0.4215561 60	24	0.0130
$^{10}_5\text{B}^{10}$	3	0.64462 22	1	0.0181
$^{11}_5\text{B}^{11}$	3/2	0.96246 10	1,23	0.0181
$^{13}_6\text{C}^{13}$	1/2	0.251430 50	10	0.0236
$^{19}_9\text{F}^{19}$	1/2	0.940930 15	18,10,23	0.0432
$^{23}_{11}\text{Na}^{23}$	3/2	0.793512 75	1,23	0.0581
$^{27}_{13}\text{Al}^{27}$	5/2	1.30283 13	1,23	0.0744
$^{31}_{15}\text{P}^{31}$	1/2	0.404835 37	1,9	0.0920
$^{35}_{17}\text{Cl}^{35}$	3/2	0.294000 93	1	0.111
$^{63}_{29}\text{Cu}^{63}$	3/2	0.795291 93	1,23,25	0.238
$^{65}_{29}\text{Cu}^{65}$	3/2	0.85185 11	1,25,23	0.238
$^{69}_{31}\text{Ga}^{69}$	3/2	0.72030 36	9	0.261
$^{71}_{31}\text{Ga}^{71}$	3/2	0.91485 33	9	0.261
$^{79}_{35}\text{Br}^{79}$	3/2	0.75177 15	23	0.309
$^{81}_{35}\text{Br}^{81}$	3/2	0.81026 11	23,1,26	0.309
$^{85}_{37}\text{Rb}^{85}$	5/2	0.48288 15	1	0.333
$^{87}_{37}\text{Rb}^{87}$	3/2	0.98154 18	1,23	0.333
$^{127}_{53}\text{I}^{127}$	5/2	1.00043 21	9,23	0.548
$^{133}_{55}\text{Cs}^{133}$	7/2	0.91792 28	1	0.577
$^{139}_{57}\text{La}^{139}$	7/2	0.988750 98	30,27	0.606
$^{203}_{81}\text{Tl}^{203}$	1/2	0.571479 45	10,28	0.977
$^{205}_{81}\text{Tl}^{205}$	1/2	0.577108 45	10,28	0.977

(V. RADIO-FREQUENCY SPECTROSCOPY)

C. PARAMAGNETIC RELAXATION

Prof. A. F. Kip

R. D. Arnold

1. Resonance Absorption

A doctoral thesis entitled "Paramagnetic Resonance Absorption in Two Cupric Salts" has been written and submitted.

2. Relaxation

Our first attempt to observe spin-spin relaxation by noting the absorption of electromagnetic energy in a paramagnetic salt, with and without an applied d-c magnetic field, produced an effect too small for quantitative measurement.

The presence of the sample salt in the resonant cavity in which the absorption takes place changed the resonant frequency of the cavity from 480 Mc to 350 Mc. A new, higher-frequency cavity has been constructed, the design similar to that described in the previous progress report but with smaller dimensions.

It may be necessary to go to lower temperatures (the first run was at room temperature) to obtain adequate sensitivity.

3. Low-Temperature Resonance

Some new experiments are being planned in conjunction with the low-temperature group. We plan to investigate resonances in a number of paramagnetic salts at liquid He<sub>2</sub> temperatures. Low temperatures will greatly decrease spin-lattice relaxation broadening and may also show up other changes in absorption spectra. We hope to investigate both copper sulfate and cesium titanous alum single crystals.

D. MICROWAVE SPECTROSCOPY

Prof. M. W. P. Strandberg

J. G. Ingersoll

Prof. G. G. Harvey

H. R. Johnson

Prof. A. G. Hill

G. W. King

J. R. Eshbach

R. B. Lawrance

B. V. Gokhale

C. C. Loomis

R. E. Hillger

M. T. Weiss

1. Germane Monochloride

Work has been discontinued on this molecule, since work elsewhere on

## (V. RADIO-FREQUENCY SPECTROSCOPY)

the same molecule is being successfully carried out. Work on tellurium in an appropriate molecule will probably replace this project.

R. E. Hillger

### 2. Ketene

Work on ketene,  $\text{H}_2\text{C}_2\text{O}$  has proceeded well since the last progress report. The three strong lines of the rotation transition  $J = 1$  to  $J = 2$ , occurring at .75 cm wavelength, have been found and their frequencies measured. With the previously measured transition  $J = 0$  to  $J = 1$  occurring at 1.5 cm, we have been able to obtain accurate values for two of the reciprocal principal moments of inertia  $b$  and  $c$ . Then, using two rather inaccurate methods, we obtained a rough estimate of the value of the deviation  $\delta$ , ( $\delta = (b-c)/(a-c)$ ) from a symmetric rotor; this appears to be roughly .001 - .005. Satellite lines, possibly from vibration interaction, were not found on the 1-2 transition, probably because of unfavorable statistics and the use of the less sensitive  $K/2$  spectroscope on straight absorption.

We plan to try to obtain the structure of ketene completely by measuring the 0-1 and 1-2 transitions of deuterio-ketene. Furthermore we hope to gain enough information to interpret the satellite lines of the 0-1 line by investigating the 2-3 line with the  $K/2$  spectroscope, and by investigating the relative intensities of the components of the 0-1 transition at room and dry ice temperatures. Finally, we may look for some of the  $\Delta K = 0$ ,  $\Delta J = 0$  transitions of high  $J$  rotational levels.

H. R. Johnson

### 3. Formaldehyde

Several  $\Delta J = 0$  lines of ordinary formaldehyde  $\text{H}_2\text{C}^{12}\text{O}^{16}$  have been newly observed in the comparatively low-frequency microwave region 8,800 to 17,000 Mc. Stark effect measurements have been made on appropriate lines with  $J$  equal to 2,7,8, and 16. It is expected that within the immediate future a total of fifteen or more lines will have been measured; twelve are already known from our work and that of Bragg and Sharbaugh (29). For each  $K_{-1}$  value several lines of different  $J$  have been measured, thus making it possible to determine quite well the form and magnitude of corrections for centrifugal distortion and zero point vibration. This should, when completed, provide a useful check on current semi-rigid-rotor theory.

In order to determine the molecular structure accurately and to check the correction theory, the isotopic molecule using  $\text{C}^{13}$  will be used.



(V. RADIO-FREQUENCY SPECTROSCOPY)

The chemistry of this appears well in hand and work with the isotopic molecule will start as soon as the measurements on ordinary formaldehyde are complete.

A paper "Microwave Spectrum of Formaldehyde" was presented before the Physical Society's anniversary meeting in Cambridge, June 18, 1949.

R. B. Lawrance

4. Oxygen

The fine structure of oxygen at low pressures, the order of a millimeter of mercury, has not been detected as yet. The program is continuing with the Zeemann detection system cleaned up. So many variables, e.g., pressure, Zeemann field strength, and modulation breadth, must be investigated that the progress will be slow until the first line is resolved and the system optimized by means of it.

5. Apparatus

Work continues on a slow sweep system and the system to work at higher than room temperatures.

References

- (1) F. Bitter, Phys. Rev. 75, 1326 (1949).
- (2) S. Millman and P. Kusch, Phys. Rev. 58, 438 (1940).
- (3) P. Kusch and S. Millman, Phys. Rev. 56, 527 (1939).
- (4) J. E. Rosenthal and G. Breit, Phys. Rev. 41, 459 (1932).
- (5) H. Kopfermann and H. Kruger, Zeits. f. Physik 103, 48J (1936).
- (6) H. Kopfermann, Kernmomente, Akad. Verlagsgesellschaft, M.B.H. p. 17, (1940)
- (7) A. Bohr, Phys. Rev. 73, 1109 (1948).
- (8) See, for example, Brookhaven National Laboratory publication 1-5, by H. H. Goldsmith and D. R. Inglis, on "Spins, Magnetic Moments, and Electric Quadrupole Moments", dated October 1, 1948.
- (9) R. V. Pound, Phys. Rev. 73, 1112 (1948).
- (10) H. Poss, Phys. Rev. 75, 600 (1949).
- (11) F. Bloch, D. Nicodemus, and H. H. Staub, Phys. Rev. 74, 1025 (1948).
- (12) W. R. Arnold and A. Roberts, Phys. Rev. 71, 878 (1947).
- (13) H. A. Thomas, R. L. Driscoll and J. A. Hipple, Phys. Rev. 75, 902 (1949).
- (14) H. Taub and P. Kusch, Phys. Rev. 75, 1481 (1949).
- (15) T. F. Wimett, R. L. E. Progress Report p. 29, July 15th, 1949.
- (16) F. Bitter, N. Alpert, D. Nagle and H. Poss, Phys. Rev. 72, 1271 (1947).
- (17) F. Bloch, E. Levinthal and M. Packard, Phys. Rev. 72, 1125 (1947).

(V. RADIO-FREQUENCY SPECTROSCOPY)

- (18) Kai Siegbahn and Gunnar Lindström, Arkiv För Fysik, Band 1 Nr 6, p 193.
- (19) A. Roberts, Phys. Rev. 72, 979 (1947).
- (20) F. Bloch, A. C. Graves, M. E. Packard and R. W. Spence, Phys. Rev. 71, 551 (1947).
- (21) H. L. Anderson and A. Novick, Phys. Rev. 71, 372 (1947).
- (22) H. L. Anderson and A. Novick, Phys. Rev. 73, 919 (1948).
- (23) J. R. Zimmerman and D. Williams, Bull. Amer. Phys. Soc. p 7, April 28, 1949.
- (24) W. C. Dickinson and T. F. Wimett, Phys. Rev. 75, 1769 (1949).
- (25) R. V. Pound, Phys. Rev. 73, 523 (1948).
- (26) R. V. Pound, Phys. Rev. 72, 1273 (1947).
- (27) W. Chambers and D. Williams, Bull. Amer. Phys. Soc. p 16, June 16, 1949.
- (28) W. G. Proctor, Phys. Rev. 75, 522 (1949).
- (29) J. K. Bragg and A. H. Sharbaugh, Phys. Rev. 75, 1774 (1949).
- (30) W. Dickinson, Nuclear Moment of La<sup>139</sup>, in manuscript.

\* \* \*