Hyperfine Structure of $Rb^{85,87}$ in the $5P$ State*

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The atomic beam resonance method has been used to investigate the hyperfine structure of the $5P$ excited states of Rb^{85} and Rb^{87}. The apparatus and method of observation were similar to those in a previous experiment on the $3P$ state of sodium. For Rb^{85} in the $5P_{\frac{3}{2}}$ state, the magnetic dipole and electric quadrupole interaction constants are, respectively, $a_{\frac{3}{2}}=25.3\pm0.2$ Mc/sec and $b=24.4\pm1.3$ Mc/sec, and for the $5P_{\frac{1}{2}}$ state, $a_{\frac{1}{2}}=120.7\pm1$ Mc/sec. For Rb^{87}, the three constants are $a_{\frac{3}{2}}=85.8\pm0.7$ Mc/sec, $b=11.8\pm0.6$ Mc/sec, and $a_{\frac{1}{2}}=409\pm4$ Mc/sec. The quadrupole moments are $Q_{85}=(0.27\pm0.02)\times10^{-24}$ cm^2 and $Q_{87}=(0.13\pm0.01)\times10^{-24}$ cm^2, which agree with the moments calculated from the hyperfine structure of these isotopes in the $6P$ state as measured by another technique.

INTRODUCTION

THIS paper deals with an application of the atomic beam resonance method[1] to the study of the hyperfine structure of Rb^{85} and Rb^{87} in the $5P$ state.[2] Since this experiment was necessarily performed with the use of higher radio-frequencies than those required in the previous experiment[3] on Na^{23}, certain important modifications were made in the method of rf generation and measurement.

Although the ratio of the electric quadrupole moments of Rb^{85} and Rb^{87} has been determined[4,5] from the spectra of the RbCl and RbF molecules, a determination of the magnitude of the quadrupole moments requires a knowledge of the wave functions of these molecules to an accuracy which is not available. The objective of this experiment was to obtain sufficient information about the hfs of these isotopes in the $5P_{\frac{1}{2}}$ and $5P_{\frac{3}{2}}$ states which, together with the more accurately known atomic wave functions, can yield a better value of the nuclear electrical quadrupole moment than can be obtained from molecular data. Added interest was given to the measurements by the results, published during the course of this experiment, on the hfs of the $6P$ state obtained by the double resonance optical method.[6] A comparison could thus be made of the magnetic dipole and electric quadrupole interaction in two atomic states with different principal quantum numbers.

APPARATUS

Since our apparatus was similar to that previously described,[3] only the modifications will be discussed.

The beam was obtained by heating a mixture of rubidium chloride and calcium shavings in the oven to a temperature of from 300°C to 350°C.

As sources of resonance radiation, two types of spectral lamps were tested: one manufactured by the Phillips Company and the other by Osram. Both lamps have as their main component a glass tube containing both rubidium and an inert gas. This discharge tube is surrounded by a vacuum jacket which keeps a uniform temperature distribution around the tube and prevents the condensation of rubidium on the walls. For the purposes of this experiment the effectiveness of the source is determined by the number of atoms it can excite to the $5P$ state, and to this end the Osram lamp was found to be superior. An improvement in the effectiveness of this source was obtained by operating it at 18.5 volts and 0.7 amp rather than at the manufacturer's rating of 10 volts and 1.5 amp. This was accomplished by increasing both the driving voltage and the ballast resistance so that the point of operation on the negative resistance characteristic of the arc moved to the desired position. Further improvement was obtained by surrounding the vacuum jacket of the discharge lamp with a water-cooling jacket. Without cooling, the number of atoms excited to the $5P$ state increased at first, reached a maximum, and then began to decrease. With cooling there was no subsequent decrease in the light effect. These phenomena are consistent with the assumption that the increased

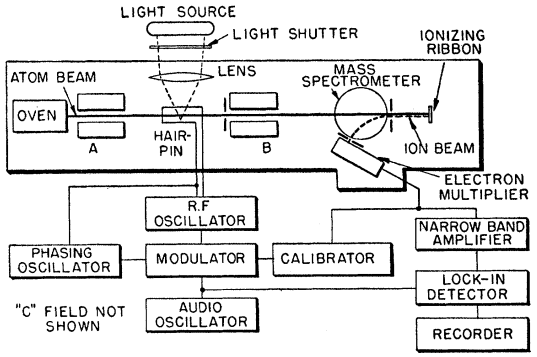

FIG. 1. Schematic drawing of the apparatus.

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† Submitted by Benjamin Senitzky in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Faculty of Pure Science, Columbia University. Now at Bell Telephone Laboratories, Murray Hill, New Jersey.

[1] I. I. Rabi, Phys. Rev. **87**, 379 (1952).
[2] Senitzky, Rabi, and Perl, Phys. Rev. **98**, 1537(A) (1955).
[3] Perl, Rabi, and Senitzky, Phys. Rev. **98**, 611 (1955).
[4] J. W. Trischka and R. Braunstein, Phys. Rev. **96**, 963 (1954).
[5] V. Hughes and L. Grabner, Phys. Rev. **79**, 314 (1950).
[6] U. Meyer-Berkhout, Z. Physik **141**, 185 (1955).

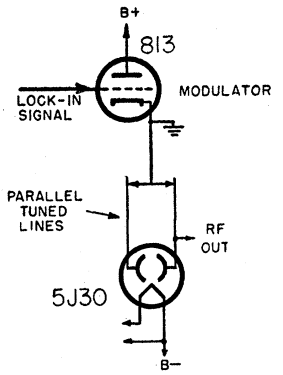

FIG. 2. Magnetron oscillator circuit schematic.

vapor pressure in the discharge tube resulting from higher temperatures leads to a greater degree of self-reversal.

The gain of the amplifier was measured by feeding a chopped dc signal of known amplitude from the calibrator (see Fig. 1) to the amplifier input. The proper relative phase of the detector and modulator was achieved in the following manner: a transition between adjacent m_F levels corresponding to the same F level in the ground state was obtained with a modulated oscillator and the resultant signal was observed on an oscilloscope. The phase of the detector was then adjusted with respect to that of the modulator. The frequency of the ground state transition was much lower than the frequencies measured in the excited state; consequently a special oscillator ("phasing oscillator" in Fig. 1), covering a range from 0.5 to 3 Mc/sec, was designed for this purpose.

At frequencies up to 180 Mc/sec the rf generation and measurements remained the same as previously described.[3] At higher frequencies, the rf power was generated by a Radar Jammer APT-4 with a frequency range from 180 to 700 Mc/sec and an rf power output of 150 to 200 watts. This oscillator, shown schematically in Fig. 2, is designed to use either of two split-anode magnetrons. For the frequency range of 180 to 400 Mc/sec a 5J30 magnetron, which feeds into an external quarter-wavelength line, is used. For the higher range of 400 to 700 Mc/sec a 5J29 magnetron (not shown in Fig. 2), which has an additional short-circuiting back loop within the tube envelope, is used. The anodes of this tube are located in the region of a voltage antinode of a half-wavelength line. The filaments of both tubes require about 35 amperes when the magnetrons are operated at low frequency and the potential difference between the anodes and filament is 1000 volts. At high frequencies the filament current of the 5J29 could be almost turned off and the filament heated by electron back bombardment. The lock-in signal is applied to the control grid of the 813 pentode which functions as a stabilizing load on the magnetron and as a modulator.

The oscillator output is connected through an attenuator to a double-stub tuner which controls the current to the hairpin. The rf power is transmitted through a type N UG-30C/U connector into the vacuum system where a length of rigid coaxial line terminates in the hairpin. The method of measuring rf current in the hairpin at frequencies below 180 Mc/sec was previously described.[3] At higher frequencies, the accuracy of this measurement becomes dependent on theoretical corrections based on electrical line lengths which are not known accurately. Consequently, a calibration procedure, which yielded the voltage in the hairpin transmission line outside the vacuum as a function of frequency for constant current in the hairpin proper, was adopted and is described below. The essential purpose of the calibration was to enable a resonance curve of considerable width to be observed at a constant rf amplitude and thus to avoid distortions arising from a variation of matrix elements over the range of observation.

The rf generation and transmission components were removed from the vacuum system and arranged as shown in Fig. 3. A small pickup loop was placed close to the hairpin and a General Radio vacuum tube voltmeter type 1800A which, according to the manufacturer's specifications, can be used to measure voltages at frequencies up to 500 Mc/sec read the induced loop voltage. Although a measurement of the absolute current in the hairpin could not be obtained because of the difficulties of calculating the mutual inductance of the hairpin and the pickup loop, the relative rf current as a function of frequency could be obtained under the assumption that the induced loop voltage is proportional to the product of frequency and hairpin current. The latter assumption is valid when, as in our case, the dimensions of the circuit are small compared to a quarter wavelength. Another voltmeter read the transmission line voltage near the double-stub tuner. To obtain a reliable calibration several precautions had to be observed. It was found necessary to shield the hairpin and pickup loop thoroughly and to measure the induced voltage as close to the pickup loop as possible. Furthermore, the oscillator had to be well isolated by an attenuator which consisted of 100 ft of RG-9/U cable and resulted in a power attenuation of 4 db at 300 Mc/sec and increased attenuation at higher frequencies. Its purpose was twofold. It kept the oscillator frequency nearly independent of the tuning position and it preferentially suppressed higher harmonics which were quite trouble-

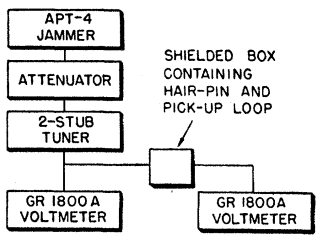

FIG. 3. Hairpin calibration arrangement.

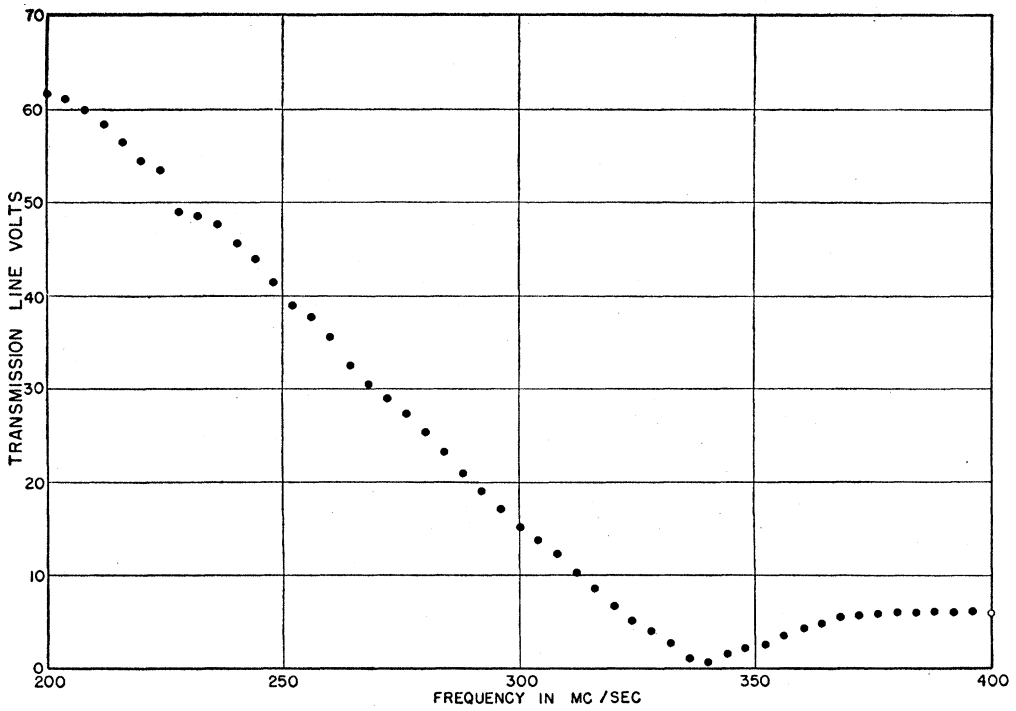

FIG. 4. Hairpin calibration curve.

some. In the region of 150 to 250 Mc/sec this filtering action was not sufficient; consequently a low pass filter (Microlab FLH 200) was placed between the oscillator and the attenuator. The calibration curve is shown in Fig. 4. To obtain a qualitative check on the above calibration procedure, the rigid coaxial line in the vacuum system and the hairpin itself can be considered to a first approximation as two series transmission lines with parameters which can be roughly calculated. The calibration curve can then be theoretically obtained and compared with the experimental curve. It was found that the general shapes of the two curves are in agreement.

DISCUSSION OF ERRORS

The transitions of interest in this experiment are those in which F changes by one in a C field which is zero. In practice, the latter condition could not be attained because of the Majorana effect. This effect, which is well known in molecular and atomic beam resonance work, occurs when the separation between adjacent m_F levels becomes small enough so that transitions between these levels can occur due to the spatially changing magnetic fields encountered by the atom in traversing the region between the two deflecting fields. The Majorana effect is undesirable in our experiment because it increases the background and decreases the signal-to-noise ratio, but it can easily be eliminated by the application of a small C field. The excited state pattern will be affected by the C field in that the resonance for the $\Delta F=\pm 1$ transition will be broadened and its maximum shifted. An exact calculation of these effects is extremely complicated and requires an exact knowledge of both the C field and rf field intensity throughout the transition region. An order of magnitude estimate was made under the assumption that the excited state m_F levels do not overlap in energy (which in this case is not a good assumption) so that the intensity of each one of the lines in the Zeeman multiplet where F changes by one can be calculated. Under this assumption, the rf changes the probability $W(c,a)$ of an atom starting in ground state (a), going through excited states (b) or (b') and then returning to the final ground state (c) by the amount[7]

$$\Delta W=[D(c,b')-D(c,b)][E(b,a)-E(b',a)]\times\frac{V_{b'b}^2}{2[V_{b'b}^2+k^2+(\omega_{b'b}-\nu)^2]},\quad(1)$$

where the D's represent the coefficients of spontaneous decay from excited state (b) or (b') to ground state (c), the E's are coefficients of absorption, the $V_{b'b}$ are the matrix elements of the rf perturbation between states (b) and (b') (in frequency units), $k=1/2\pi\tau$ where τ is the lifetime of the excited state, and ν and $\omega_{b'b}$ are the impressed and resonant frequencies respectively. The intensity of the signal arising from a particular transition in the multiplet from state (b) to (b') is obtained by summing (1) over all initial and final states which differ in their moments in the deflect-

[7] See, reference 3. Also R. Serber, Special Technical Report of Columbia Radiation Laboratory (to be published).

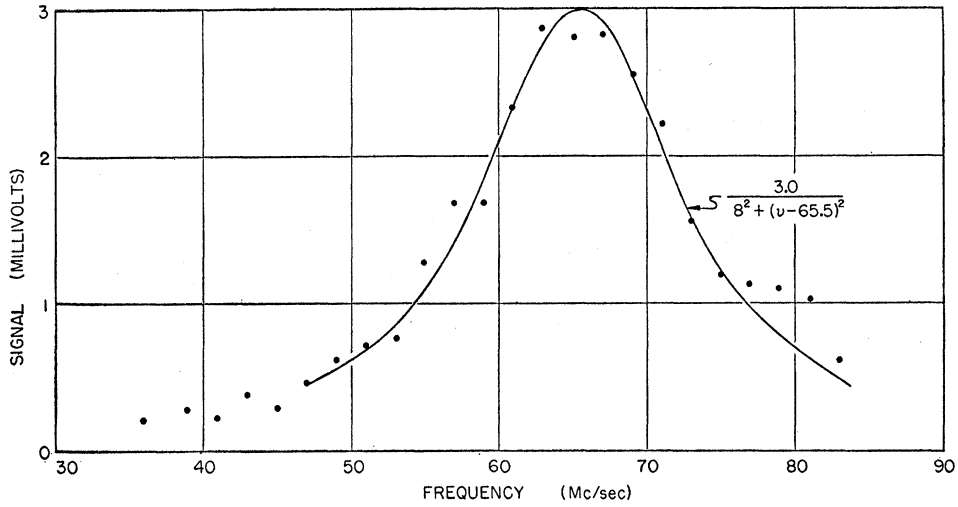

FIG. 5. The $F=3$ to $F=2$ transition of Rb^{85} in the $5P_{\frac{3}{2}}$ state.

ing fields. This procedure is repeated for all transitions in the multiplet and the shift of its center of gravity from the zero field position estimated. This shift for our experiment was estimated to be 0.1 Mc/sec upwards in frequency at the typical operating field of one gauss. This error is negligible compared to other errors. It is unlikely that a complete theory which allows for the interference of the probability amplitudes would predict a shift of greater magnitude although it might give some structure in the close neighborhood to the center of the resonance.

The perturbation of the energy levels by the rf field can be calculated without the assumptions made above.[7] This is done at zero C field and with the axis of quantization along the direction of the rf field. The change in energy $\Delta\omega_b$ (in frequency units) of excited state level (b) is given by

$$\Delta\omega_b=-\frac{1}{4}\sum_{b'}\left[\frac{V_{b'b}^2}{\omega_{b'b}-\nu}+\frac{V_{b'b}^2}{\omega_{b'b}+\nu}\right], \tag{2}$$

where $\omega_{b'b}$ is the Bohr frequency between levels (b) and (b') and the summation is to be carried out over all levels with the same m_F omitting the terms where the denominator approaches zero. For our case, the shift in transition frequencies due to this effect is less than 0.1 Mc/sec so that this effect is also negligible compared to the other errors.

The two important factors which were considered in assigning the uncertainty to the final results of the measurements were the statistical uncertainty and the uncertainty arising from the current calibration technique.

TABLE I. Data on the 65.0-Mc/sec resonance.

Run	Resonant freq. (Mc/sec)	Half-width (Mc/sec)	rf current (amp)
1	65.0	9.0	2.0
2	64.5	9.5	2.0
3	65.0	9.5	2.0
4	64.8	8.5	2.0
5	65.5	8.0	1.2

EXPERIMENTAL RESULTS

The line of lowest frequency which was observed and which will be later shown to be assignable to the Rb^{85} spectrum was at 65.0±0.75 Mc/sec. Five runs were taken on this resonance and the results are given in Table I. A typical resonance is plotted in Fig. 5 where the signal is given in millivolts measured at the recorder. To determine the resonant frequency it was assumed that the resonance could be represented by a curve of the form

$$S=A\,V^2/[V^2+k^2+(\omega-\nu)^2], \tag{3}$$

where S is the signal, V is a constant proportional to the amplitude of the rf field, A is a constant proportional to the gain of the detection system, and ω is the resonant frequency. It can be seen from Fig. 5 that the data can be fairly well fitted with a curve which has the frequency dependence shown in Eq. (3). The dependence on rf amplitude of the above function was checked by setting the frequency at 65 Mc/sec and plotting the signal *vs* the rf current. If Eq. (3) is valid, it should be possible to fit the data with the function

$$S=A/[1+(K/I)^2], \tag{4}$$

where A and K are two constants to be determined from the best fit and I is the rf current. K is a constant which is proportional to the natural line width and A has been previously defined. The resultant data are shown in Fig. 6. The range of the rf currents is limited at the low end by the poor signal-to-noise ratio and at the high end by the available output of the oscillator. Although there is an indication that the rf dependence is of the form given by Eq. (4), not enough data are available for a thorough investigation of this question.

A resonance was found at 120.8±1.5 Mc/sec and

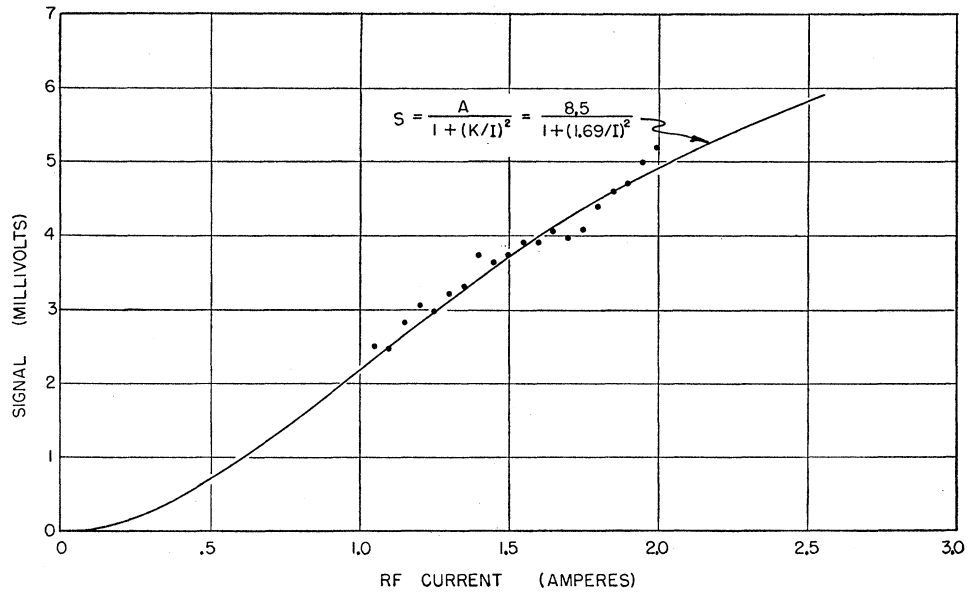

FIG. 6. Signal *vs* rf current at 65.0 Mc.

will later be shown to be assignable to the Rb^{85} spectrum. The data are shown in Fig. 7. It was found that the center of the line could be determined from the individual points to the accuracy desired so that a curve is not drawn through the data in this case. The intensity of this line was approximately twice that of the 65.0-Mc/sec resonance. Three runs were taken on this resonance and the results are given in Table II.

A resonance was found at 265±3 Mc/sec and assigned to the Rb^{87} spectrum (based on the use of an enriched sample of this isotope). The data shown in Fig. 8 were taken with a sample where the two isotopes were in the natural abundance ratio of Rb^{85} to Rb^{87} of 4 to 1. As can be seen, the signal-to-noise factor is less than for the Rb^{85} lines, which is as expected, since the signal-to-noise factors estimated on the basis of abundance alone should be 16 to 1. This results from the fact that the same abundance ratio exists in the light source as in the beam and the background remains the same for both isotopes.

A resonance with a very weak intensity was found at 362±3 Mc/sec and assigned to the $5P_{\frac{1}{2}}$ state of Rb^{85}. The frequency of this line is in agreement with that predicted from optical measurements of the hyperfine structure of the Rb^{87} $5P_{\frac{1}{2}}$ state[8] and the known ratio of a_{85}/a_{87}. The poor signal-to-noise ratio encountered in our experiment made the measurement of this frequency the most difficult of any undertaken. Five runs with the results of a typical one shown in Fig. 9, are listed in Table III. To establish the reality of this resonance, it was also observed with the C field set at 33 gauss. Although the pattern seemed to be shifted, conclusive data on the Zeeman effect were not obtained. A further check was to take two runs with an enriched sample containing over 95% of Rb^{87} with a typical result shown in Fig. 10. The absence of a resonance here is consistent with the assignment of the line to the Rb^{85} spectrum.

CALCULATION OF INTERACTION CONSTANTS

If the electronic orbital angular momentum is designated by L, the total electronic angular momentum by J, and the nuclear spin by I, the contribution of the nuclear magnetic dipole and electric quadrupole

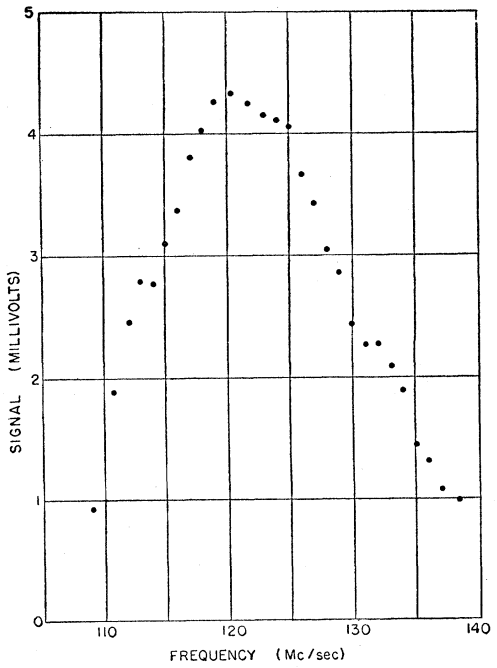

FIG. 7. The $F=4$ to $F=3$ transition of Rb^{85} in the $5P_{\frac{1}{2}}$ state.

[8] H. Kopferman and H. Krüger, Z. Physik **103**, 485 (1936).

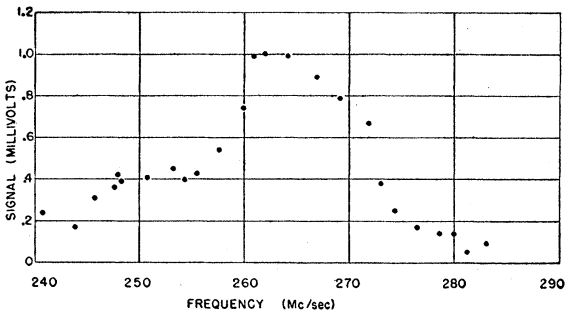

FIG. 8. The $F=3$ to $F=2$ transition of Rb^{87} in the $5P_{\frac{3}{2}}$ state.

interaction to the atomic energy level $E(\text{hfs})$ may be written as[9]

$$E(\text{hfs})=ha\frac{C}{2}+hb\left[\frac{\frac{3}{8}C(C+1)-\frac{1}{2}I(I+1)J(J+1)}{IJ(2I-1)(2J-1)}\right], \quad (5)$$

where $C=F(F+1)-I(I+1)-J(J+1)$.

The primary quantities which are determined in our experiment are the interaction constants a and b. They may also be calculated theoretically as

$$ha=\frac{2\mu_I\mu_0}{I}\frac{L(L+1)}{J(J+1)}\mathfrak{F}\left\langle\frac{1}{r^3}\right\rangle_{\text{Av}} \quad (6)$$

and

$$hb=e^2\frac{2L}{2L+3}R\left\langle\frac{1}{r^3}\right\rangle_{\text{Av}}Q, \quad (7)$$

where μ_I is the nuclear magnetic moment in emu, μ_0 is the Bohr magneton, and $\mathfrak{F}$ and R are relativistic corrections. An approximate value of $\langle 1/r^3\rangle_{\text{Av}}$ can be obtained from the fine structure splitting according to a method of Barnes and Smith,[10] from which a may be calculated by use of Eq. (6).

The a and b values can be determined experimentally from the hyperfine spectrum where the transition

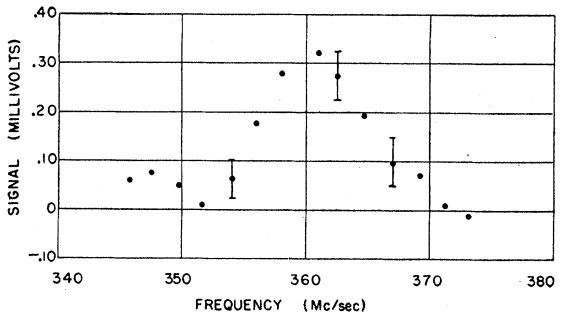

FIG. 9. The $F=3$ to $F=2$ transition of Rb^{85} in the $5P_{\frac{1}{2}}$ state.

[9] H. B. G. Casimir, *On the Interaction between Atomic Nuclei and Electrons* (Teylers Tweede Genootschap, Haarlem, 1936).
[10] R. G. Barnes and W. V. Smith, Phys. Rev. **93**, 95 (1954).

frequencies are found from Eq. (5) to be

Rb^{87} ($I=3/2$)		Rb^{85} ($I=5/2$)	
Transition	Frequency	Transition	Frequency
$F=3$ to $F=2$	$3a+b$	$F=4$ to $F=3$	$4a+(4/5)b$
$F=2$ to $F=1$	$2a-b$	$F=3$ to $F=2$	$3a-(9/20)b$
$F=1$ to $F=0$	$a-b$	$F=2$ to $F=1$	$2a-(4/5)b$

In the Rb^{85} spectrum, the 120.8 Mc/sec resonance is assigned to the $F=4$ to $F=3$ transition and the 65.0 Mc/sec resonance is assigned to the $F=3$ to $F=2$ transition to yield values of $a=25.3\pm0.2$ Mc/sec and $b=24.4\pm1.3$ Mc/sec. The theoretical value of a calculated according to the method mentioned above is found to be 24.3 Mc/sec. Furthermore from the known ratio of nuclear moments (see below) the frequency of the $F=3$ to $F=2$ transition in the Rb^{87} spectrum can be computed and is found to agree, to within the limits of error, with the experimental value of 265 ± 3 Mc/sec. This agreement serves both to identify the observed line as the $F=3$ to $F=2$ transition

TABLE II. Data on the 120.8-Mc/sec resonance.

Run	Resonant freq. (Mc/sec)
1	119.6
2	121.4
3	121.3

TABLE III. Data on the 362-Mc/sec resonance.

Run	Resonant freq. (Mc/sec)
1	364
2	362
3	363
4	361
5	360

of Rb^{87} and to give added confirmation to the assignment of the lines to the Rb^{85} spectrum. Other assignments of the two Rb^{85} resonances yield a values which are above 30 Mc/sec and are inconsistent with any assignment of the Rb^{87} resonance. From the ratio of the a's (obtained from the known nuclear moments and spins[11]) of $a_{85}/a_{87}=0.295$ and the ratio[4] $b_{85}/b_{87}=2.07$, the values of the interaction constants of Rb^{87} in the $5P_{\frac{3}{2}}$ state are

$$a=85.8\pm0.7 \text{ Mc/sec} \quad \text{and} \quad b=11.8\pm0.6 \text{ Mc/sec}.$$

The ratio of the interaction constants in the $5P_{\frac{3}{2}}$ states measured in this experiment to the interaction constants of the $6P_{\frac{3}{2}}$ states[6] is

$$a_{5P}/a_{6P}=3.1\pm0.035 \quad \text{and} \quad b_{5P}/b_{6P}=2.9\pm0.2.$$

The ratio of $\langle 1/r^3\rangle_{\text{Av}}$ as computed[10] for the two states is found to be 3.08, in agreement with the above. In comparing this ratio of $\langle 1/r^3\rangle_{\text{Av}}$ with the experimental

[11] P. A. Kusch and S. Millman, Phys. Rev. **56**, 527 (1939).

ratios, the effect of the inner core of electrons[12] was not considered. This effect was computed for the two states in question for the b ratio by Sternheimer[13] and the results indicate that the theoretical ratio is not significantly changed from that given in the foregoing.

In the $P_{\frac{1}{2}}$ state of Rb^{85}, the interval between $F=3$ and $F=2$ is $3a$. From the resonance at 362 ± 3 Mc/sec the $a_{\frac{1}{2}}$ is found to be 120.7 ± 1 Mc/sec. If it is assumed that the nuclear magnetic dipole moment interacts only with the valence electron, the ratio $a_{\frac{1}{2}}/a_{\frac{3}{2}}=5.59$ whereas the experimental ratio is 4.8 ± 0.05. One possible explanation[14] for this discrepancy is the effect of excited 4-shell configurations. The resultant perturbation of the magnetic fields at the nucleus is somewhat different for the $P_{\frac{1}{2}}$ and $P_{\frac{3}{2}}$ states, and could account for the difference between the observed value of $a_{\frac{1}{2}}/a_{\frac{3}{2}}$ and that expected theoretically for a single $5p$ electron outside a closed shell.

The quadrupole moment computed from the $a_{\frac{3}{2}}$ without Sternheimer shielding corrections is found to be[15]

$$Q_{85}=\frac{\mu_I\mu_0}{e^2}\frac{(L+1)(2L+3)}{IJ(J+1)}\frac{\mathfrak{F}}{R}\frac{b}{a}$$

$$=(0.27\pm0.02)\times10^{-24}\text{ cm}^2,$$

$$Q_{87}=(0.13\pm0.01)\times10^{-24}\text{ cm}^2,$$

[12] R. M. Sternheimer, Phys. Rev. **95**, 737 (1954).
[13] R. M. Sternheimer (private communication).
[14] M. Phillips, following paper [Phys. Rev. **103**, 322 (1956)].
[15] Davis, Feld, Zabel, and Zacharias, Phys. Rev. **76**, 1076 (1949).

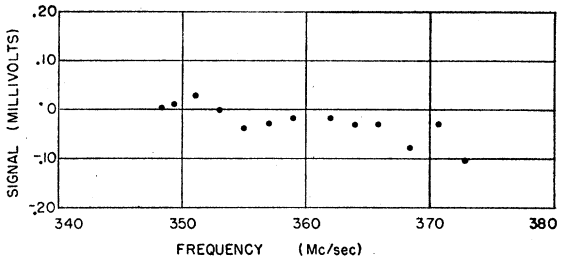

FIG. 10. The frequency region of the Rb^{85} $5P_{\frac{1}{2}}$ state resonance with an enriched sample of Rb^{87} in the oven.

in agreement with the results of the double-resonance optical method.[6] Application of Sternheimer's correction[13] of 0.88 to the above values yields

$$Q_{85}=(0.24\pm0.02)\times10^{-24}\text{ cm}^2,$$

$$Q_{87}=(0.12\pm0.01)\times10^{-24}\text{ cm}^2.$$

These values can be compared to the theoretical values obtained from the Rainwater model of the distorted nuclear core[16] of

$$Q_{85}=0.33\times10^{-24}\text{ cm}^2,$$

$$Q_{87}=0.16\times10^{-24}\text{ cm}^2.$$

ACKNOWLEDGMENTS

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[16] R. van Wageningen and J. de Boer, Physica **18**, 369 (1952).