Advanced Laboratory Experiments on Optical Pumping of Rubidium Atoms—Part I: Magnetic Resonance

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The construction and operation of a simple apparatus for optical pumping experiments using rubidium vapor are described. The experiments are intended to involve students with the concepts of resonance, atomic orientation, nuclear magnetic moments, and the precession of atomic magnetic moments. An outline of the elementary theory is included.

INTRODUCTION

SUGGESTIONS for the use of optical pumping experiments in the undergraduate laboratory have appeared previously.[1,2] The present two-part article[3] describes the construction and operation of a simple apparatus in greater detail and extends the range of student experiments. The experiments illustrate the need to invoke nuclear magnetic moments, the concepts of atomic orientation and polarization, and the precession of atomic magnetic moments.

In thermal equilibrium the relative distribution of atoms between any two energy states is determined by the Boltzmann factor $(N_2/N_1)=\exp(-\Delta E/kT)$, where $\Delta E=E_2-E_1$, E_1, and E_2 are the energies of the two states, and N_1 and N_2 are the numbers of atoms in these states. For values of $\Delta E/h$ corresponding to radio frequencies and for temperatures in the vicinity of 300°K, the population excess in the lower level is extremely small. Optical pumping is a process in which a marked change from the thermal equilibrium population of the energy levels is induced by an optical method. Pumping of atoms from the ground state into excited states is achieved by irradiation of the atoms with characteristic optical resonance radiation. The elegance of the optical pumping technique results partly because of the simple method of detection: monitoring the transmission of the resonant optical radiation.

The development of the optical pumping technique was initiated in 1949 by Brossel and Kastler.[4,5] Since then, methods for optical pumping have been improved and applications of many kinds have been described. General treatments of the subject can be found in review articles by Benumof,[2] Skrotskii and Izyumova,[6] and Carver.[7] As applied to Rb atoms the theory in outline is as follows: Stable rubidium exists in two isotopic forms ^{85}Rb ($I=5/2$) and ^{87}Rb ($I=3/2$). Though our discussion is limited to the $I=3/2$ isotope, optical pumping of the $I=5/2$ isotope can be explained in a very similar way.

The energy diagram for the ground state and the first excited state of ^{87}Rb is shown in Fig. 1. In the absence of external magnetic fields, Rb atoms exhibit a spectrum corresponding to the hyperfine structure. When the Rb atoms are subjected to a weak magnetic field (such as the earth's) further level splitting occurs, and the atomic spectrum exhibits the anomalous Zeemen effect. The additional energy splitting is given by $\Delta E=g_F\mu_0H_0$, where g_F is the Landé g-factor, μ_0 the Bohr magneton, and H_0 the external magnetic field. Since $\Delta E\ll kT$ the populations of the Zeeman sublevels are nearly equal.

When ^{87}Rb vapor, at low pressure, is irradiated with filtered 7948 Å Rb-resonance radiation (see Fig. 1) electric dipole transitions from $2S_{\frac{1}{2}}\rightarrow 2P_{\frac{1}{2}}$ but not $^2S_{\frac{1}{2}}\rightarrow {}^2P_{\frac{3}{2}}$ are induced (an interference filter is used to eliminate the D_2 line which could provide an escape route through a $^2P_{\frac{3}{2}}$ state for the pumped atoms). If circularly polarized resonance light corresponding to $\Delta M_F=+1$ is used, upon each upward transition the z component

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1 R. L. deZafra, Am. J. Phys. 28, 646 (1960).

2 R. Benumof, Am. J. Phys. 33, 151 (1965).

3 Part II is the following paper in this issue.

4 J. Brossel and A. Kastler, Compt. Rend. 229, 1213 (1949).

5 A. Kastler, J. Phys. Radium 11, 255 (1950).

6 G. V. Skrotskii and T. G. Izyumova, Sov. Phys. Usp. Fiz. Nauk 73, 423 (1961) [English transl.: Soviet Phys.—Usp. 4, 177 (1961)].

7 T. Carver, Science 141, 599 (1963).

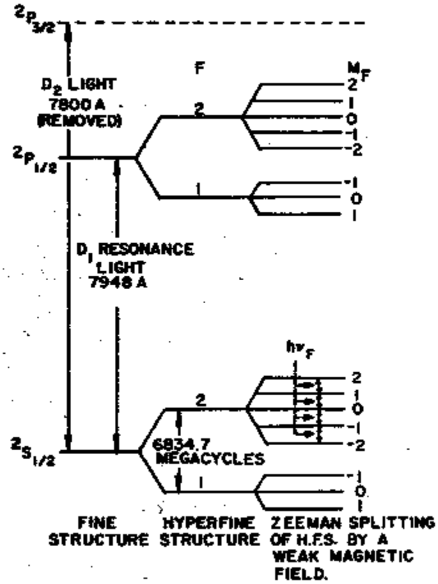

FIG. 1. A schematic representation (not to scale) of the splitting of the lowest atomic energy levels of ^{87}Rb in a weak magnetic field. The D_2 line is removed by a filter from the irradiating beam.

of the atom's angular momentum (i.e., its M_F quantum number) increases by one unit. For instance, consider the $F=1$, $M_F=0$ sublevel of the $^2S_{\frac{1}{2}}$ state. Upon irradiation it is excited to the $^2P_{\frac{1}{2}}$, $F=1$ or 2, $M_F=+1$ state. In returning to the ground state, the atoms emit either circularly or linearly polarized light, corresponding to $\Delta M_F=\pm 1$ or $\Delta M_F=0$ transitions. Thus, some of the atoms in our example return to $F=2$, $M_F=2$ sublevel in the $^2S_{\frac{1}{2}}$ state. Atoms in the $M_F=2$ state cannot absorb another unit of angular momentum, and so, remain unaffected by further radiation. The rest of the atoms in the lower sublevels can continue to absorb circularly polarized resonance light, until they gradually accumulate in the $M_F=2$ state.

Were there no relaxation effects, in time all the atoms would have been pumped to the $M_F=2$ state, bringing about complete orientation of the sample, which then would be transparent to the incident radiation. Even though complete orientation is never attained experimentally, we assume in the following that, in good approximation, all ^{87}Rb atoms are pumped into the $F=2$ state and remain there.

The relation between the absorption coefficient $K(P_Z)$ and the electronic polarization P_Z is

$$K(P_Z)=K_0(1-P_Z),$$

where $K_0=K(P_Z=0)$, and $P_Z=(N_{\frac{1}{2}}-N_{-\frac{1}{2}})/(N_{\frac{1}{2}}+N_{-\frac{1}{2}})$ with the N's referring to the numbers of atoms in $M_s=\pm\frac{1}{2}$. This is easily verified for $I=0$ and $J=\frac{1}{2}$. For the experimentally realized case of irradiation with spectral components of sufficient width, the optical absorption depends only on the electronic polarization of the atom. Therefore, this formula is also valid for $I\neq 0$. In the experimentally interesting case of polarization approaching unity and weak rf field (with $F=2$ as mentioned above), on account of the parallelism of the vectors **I** and **S** we can write, in general, for the electron polarization

$$P_Z=\sum M_F N(M_F)/2\sum N(M_F),$$

where $N(M_F)$ is the number of atoms in the particular M_F state (Fig. 1) and the summation is over all values of M_F for $F=2$.

In order to detect the polarization of the sample a radio-frequency field H_1 is used to perturb the oriented atoms. According to the selection rules ($\Delta F=0$, $\Delta M_F=\pm 1$) magnetic dipole transitions can be induced among the sublevels M_F of the $^2S_{\frac{1}{2}}$ state. Hence a radio-frequency field H_1 is applied in a direction normal to H_0 at the frequency ν_F given by

$$\nu_F=\Delta E/h=g_F\mu_0 H_0/h.$$

The magnetic dipole transitions tend to equalize the Zeeman sublevel populations, hence destroying the polarization and enabling further absorption of light. A reduction of the light intensity from $I(P)$, for a polarized sample to $I(0)$, for one in statistical equilibrium occurs when the H_1 oscillator frequency ν approaches the Zeeman frequency ν_F. If the oscillator frequency is swept through this value, the transmitted light intensity then passes through a minimum. Alternatively, the H_0 field may be modulated to sweep the atoms through the resonance frequency. This latter method was adopted for experiments described here, since it is more easily accomplished.

CONSTRUCTION OF APPARATUS

The experimental apparatus was designed to be compact and easy to take apart, so that students can look at its parts and, also, for convenient use for magnetic field monitoring. It consists of two main parts: optical equipment including the Rb lamp, absorption cell, etc., which are housed in a phenolic tube called the *outer shell*; and electronic equipment.[8]

The phenolic tube (2 in. i.d. and 14 in. in length) houses a Rb spectral lamp, a lens, an interference filter, a circular polarizer, a Rb absorption cell, and a photo detector, as shown in Fig. 2. Power for the Rb lamp and the H_1 and H_0 modulation fields is led into the tube through a six prong Jones plug. The Jones plug is cemented to a 2-in.-diam fiber disk which can be mounted by means of screws at the bottom of the tube. To lead power from the Jones plug connection at the bottom of the tube to the H_1 and H_0 modulation windings, wires go through a small hole or slit cut near the disk. The Rb lamp[9] is fastened to the tube by a screw through the tube to a tapped hole at the shield of the Rb lamp.

The photo sensitive element is a 0.2-in.-square solar cell. An optical system is needed to converge the light on it. A convex lens of 3 in. focal length and 2 in. diam, and a concave mirror[10] are used. The lens is cemented to the end of a thin-walled fiber tube of standard 2 in. o.d. which can slide easily into the outer shell, and can be supported by the Rb lamp. The length of the 2 in.-o.d. tube, which supports the lens, is cut to 3 in. so that the Rb bulb is at the focal point of the lens.

A mask in the form of a thin disk with a hole is placed just in front of the Rb lamp (Fig. 2). It prevents light reflected by the mirror back of the lamp from entering the absorption cell. Since most of this reflected light would not be parallel to the H_0 field it would cause a broadening of the resonance observed on applying the H_1 field. Although the mask cuts down the intensity of the signal, the line width is narrowed considerably.

A 2 in. interference filter,[11] the transmission of which is peaked at 7948 Å (the D_1 line), serves to eliminate the D_2 Rb spectral line. In order to be able to slide the filter easily in the tube, it is

[8] A complete set of the shop and electrical drawings is available on request to the Department of Physics, University of Washington, Seattle, Washington.

[9] The research rubidium spectral lamp X49-609, manufactured by Varian Associates, Palo Alto, California, is used.

[10] The lens and mirror were purchased from Edmund Scientific Co., Barrington, N. J.

[11] Obtained from Spectrolab, Inc., N. Hollywood, California.

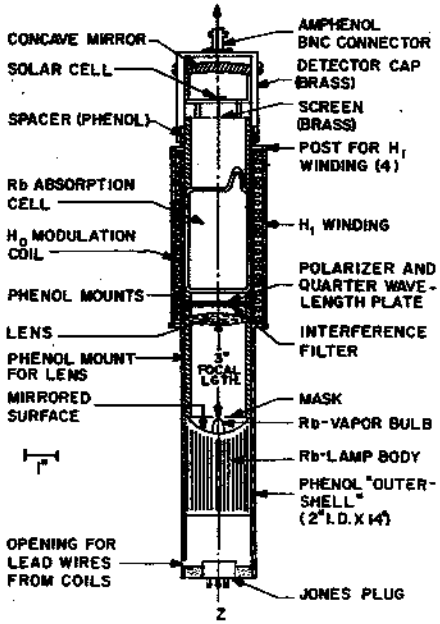

FIG. 2. The arrangement of the optical components in the fiber tube. The polarizer and quarter wave-length plate are mounted together, but the polarizer must be nearer the Rb lamp.

cemented to a thin walled ½ in. long section of the 2 in.-o.d. tubing.

The linear polarizer and quarter wavelength plate which give the light its circular polarization, can be purchased as one unit.[12] This combination is mounted in the same manner as the interference filter. Care should be taken to mount the polarizer side nearer the lamp than the quarter wavelength plate. The Rb absorption cell has a 2 in. o.d. and a 3 in. length. (Figure 2 shows the tip at its end where the cell was sealed off after preparation.) It contains a drop of purified Rb and 50 mm of neon as a buffer gas, which increases the degree of orientation.[13–15] The procedure for preparing the absorption cell is also found in Refs. 13–15.

The H_1 and H_0 modulation coils are wound on the outside of the outer shell. Since uniformity of the fields is essential, the coils are about twice the length of the absorption cell. The H_0 modulation field should be parallel to the earth's field. Hence the tube should be mounted parallel to

[12] This also was obtained from Varian Associates.

[13] J. Brossel, J. Margerie, and A. Kastler, Compt. Rend. 241, 865 (1955).

[14] W. Franzen, Phys. Rev. 115, 850 (1959).

[15] W. E. Bell, A. L. Bloom, and J. Lynch, Rev. Sci. Instr. 32, 688 (1961).

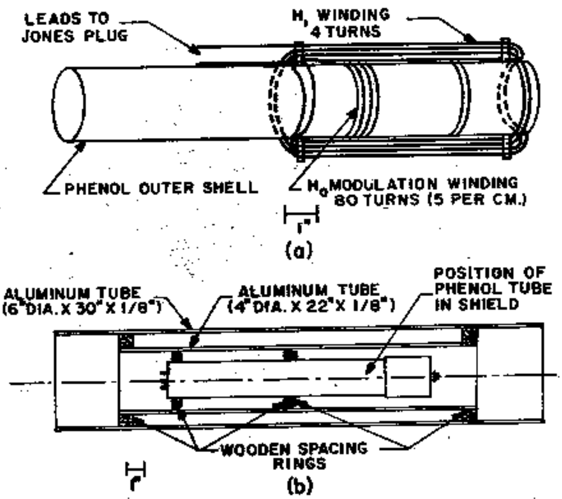

FIG. 3. (a) The arrangement of the magnetic field windings on the fiber tube. The H_0 modulation field is along the tube axis and the H_1 field across the axis. (b) The arrangement of the aluminum shielding tubes, and the location in them of the phenol tube containing the optical parts.

the earth's field. A groove with a pitch of five turns per cm is cut into the tube for this winding, which is approximately 80 turns of #20 plastic covered wire. The H_1 field must be perpendicular to the H_0 modulation field. Hence, four posts are mounted on the outer shell and four turns of #20 plastic covered wire wound around them as shown in Fig. 3(a) to produce the H_1 field.

The detection element is housed in a brass cap of 4 in. length (Fig. 4) which fits tightly over the fiber tube. The detector cap consists of a concave mirror 54 mm in diameter and 24 mm focal length, a solar cell,[16] and a screen. At the back of the detector cap is mounted an Amphenol BNC connector. The mirror is cemented to a brass ring $\frac{3}{8}$ in. long which can slide easily in and out of the cap, and can be fastened to it by means of screws at the side near its end plate. A strip of copper bent in the form of a **U** is cemented to the side of the mirror so that the copper strip is a focal length away from the mirror and its center at the focal point. To shield the solar cell from pickup, a piece of $\frac{1}{16}$-in. mesh brass screen 2 in. in diameter is mounted by means of two screws on the copper strip. The solar cell is then mounted on the strip with glue or tape and its leads are connected to the BNC connector. Care should be taken throughout the construction not to drop any magnetic particles into the tube housing the optical parts.

The constant current supply used for the Rb lamp is essentially the one described by Bell, Bloom, and Lynch,[15] with the following minor modifications. (1) Instead of grounding one side of the heater filament at the supply chassis, the heater is grounded at the Rb lamp. It is found that this minimizes 60 cps pickup. (2) To eliminate high-frequency feed back oscillation, 50 Ω resistors are inserted before the two plates and control grids. The modulation field is supplied by the sawtooth output from the oscilloscope. It is connected as shown in Fig. 5(a). This circuit is built into the constant current power supply. The H_1 field is supplied by a radio-frequency oscillator which is connected in series with a 100 Ω leak-in resistor to the H_1 coils. Power is led to the lamp and coils through a five conductor cable.

The signal to noise ratio can be improved if: (1) a double-shielded cable is used to transfer the signal from the detection cap to the scope; and (2) the solar cell, which in operation develops a bias potential of $\sim$0.2 V, is biased to an optimum operating potential. This improves the signal to noise ratio by a factor of four, and increases the absolute size of the signal. The biasing circuit, shown in Fig. 5(b), is enclosed in a grounded metal box. The signal is displayed on a Tektronix oscilloscope with a Type E high-gain preamplifier.

To eliminate modulation by pickup signals, the optical section is placed inside two concentric aluminum cylinders as indicated in Fig. 3(b). The whole is fastened to a mount which provides both horizontal and vertical rotation. The tube therefore can be aligned parallel to the earth's

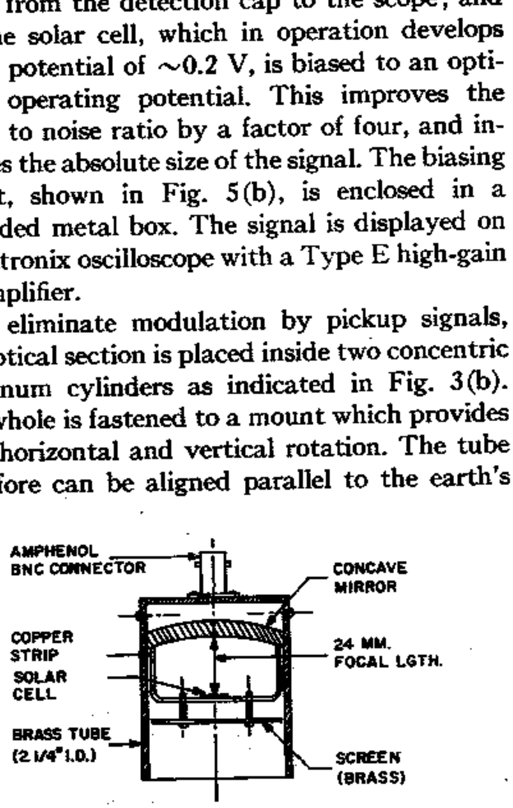

FIG. 4. Details of the detector mounting. The concave mirror has notches in its edge through which the leads from the solar cell to the BNC connector can go.

[16] Hoffman Electronics, Evanston, Ill. #55C, 0.2 in. × 0.2 in.

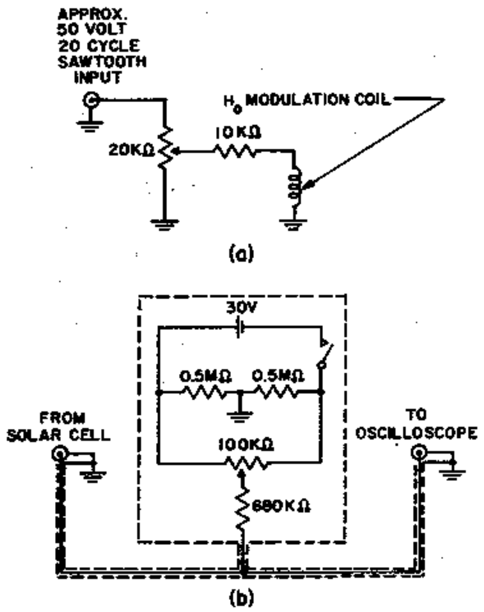

FIG. 5. (a) Connections for the H_0 modulation. These are mounted in the same chassis as the constant current power supply. (b) The circuit used to bias the solar cell to the optimum potential.

magnetic field. The outer aluminum tube is of 6 in. diam and 30 in. long, and the inner is of 4 in. diam and 22 in. long.

EXPERIMENTAL PROCEDURE

Figure 6 is a block diagram of the complete apparatus. The lines connecting the instrument to the electronic equipment should be long enough to remove the optical section from the influence of any magnetic fields in the electronic sections. With the aid of a magnetic compass and dip needle the direction of the earth's magnetic field is found and the aluminum shield with the optical tube inside is set parallel to that direction.

The Rb lamp should be given about 30 min to warm up. The absorption cell should contain enough rubidium vapor at room temperature to give a good resonance signal, but in case the signal is weak, the absorption cell can be heated gently by blowing warm air from a hair dryer through the aluminum tubes to increase the vapor density.

In determining the frequency to be used for the H_0 modulating field it should be born in mind that the period of the modulation must be longer than the optical pumping time; otherwise the modulation field prevents saturation of the pumping process and reduces the maximum orientation. It is found convenient to use a sweep time of 50 msec full scale. The modulation amplitude is turned up to about two-thirds of its full range. The H_1 amplitude is adjusted to provide about 20 mA to the H_1 coil.

For the student not familiar with the idea of a nuclear angular momentum, the following procedure is instructive. From the Zeeman splitting formula, if we assume for the moment that there is no nuclear angular momentum, $\nu_J=\gamma_J H_0$, where $\gamma_J(=g_J\mu_0/h)$ is found to be 2.8 Mc/sec/G. Since the earth's field is roughly 0.5 G, $\nu_J\sim 1.4$ Mc/sec. When the H_1 oscillator frequency is moved through this frequency, however, no resonance is observed. By extending the search to lower frequencies two resonances are found near $\nu_F^{\mathrm{I}}=0.35$ Mc/sec and $\nu_F^{\mathrm{II}}=0.23$ Mc/sec. To measure the resonance frequency precisely, the following procedure is used: the three independent variables, H_1 amplitude, H_0 modulation intensity, and frequency are adjusted to give the narrowest signal with the maximum amplitude. The H_1 amplitude is adjusted so that further power increase broadens the observed resonance. The H_0 modulation intensity and frequency are adjusted to give a large signal.

Precise measurement shows the ratio $\nu_F^{\mathrm{I}}/\nu_F^{\mathrm{II}}$ to be very nearly 3/2. The relative intensity of the two resonances is found to be approximately $I^{\mathrm{I}}/I^{\mathrm{II}}=1/3$. Comparison with the ratio of the known abundances $^{87}Rb/^{85}Rb=28\%/72\%$ sug-

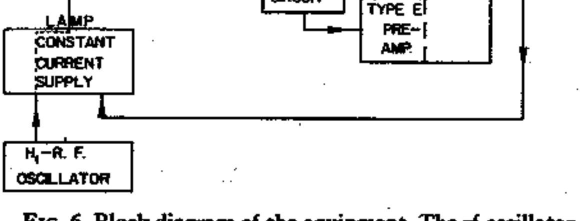

FIG. 6. Block diagram of the equipment. The rf oscillator used was a General Radio 1330A bridge oscillator, but any good oscillator covering the range 100 kc/sec to 1500 kc/sec and furnishing about 1 V across 50 Ω will do.

FIG. 7. Photo of oscilloscope trace of the ^{85}Rb resonance. The width at half maximum corresponds to about 1.3 kc/sec. Sweep time is 50 msec.

gests assigning $\nu_F{}^{\mathrm{I}}$ to ^{87}Rb and $\nu_F{}^{\mathrm{II}}$ to ^{85}Rb. The fact that two frequencies are observed and that each is much less than 1.4 Mc/sec is explained by the presence of two isotopes with two different nuclear angular momenta. Inspection of the Landé g factor formula shows that γ_J has to be replaced by γ_F to give $\nu_F=\gamma_F H_0$. For the ground state $\gamma_F=\gamma_J/(2I+1)$. To find the nuclear angular momenta which correspond to the two isotopes, the values of $2I+1$ for the lowest possible I values are:

I	0	1/2	1	3/2	2	5/2	3	7/2	4	9/2	5
$2I+1$	1	2	3	4	5	6	7	8	9	10	11

Since the observed frequencies are in the ratio 2/3, the following pairs of I values are consistent with the data: (1/2, 1), (3/2, 5/2), (5/2, 4). With $\gamma_J=2.800$ Mc/sec/G these nuclear momentum values lead to frequency pairs in the earth's field of (0.7 Mc/sec, 0.47 Mc/sec), (0.35 Mc/sec, 0.23 Mc/sec), (0.23 Mc/sec, 0.14 Mc/sec). In conjunction with the measured intensities the evidence is clearly in favor of $^{87}I=3/2$ and $^{85}I=5/2$.

Figure 7 shows a photograph of the oscilloscope trace of the frequency resonance for ^{85} Rb. It was taken by one of the students in our advanced laboratory. He found the resonances at 0.2155 Mc/sec for ^{85}Rb and at 0.324 Mc/sec for ^{87}Rb. These have the ratio of 0.665 instead of 2/3, which indicates how accurately the resonance minimum can be determined. Using the relation $\nu_F=2.800\ H_0/(2I+1)$, with $\nu_F=0.2155$ Mc/sec and $I=5/2$, the magnetic field in our laboratory is calculated to be 0.462 G. The line width of the curve shown in Fig. 7 was measured by the student to be 1.3 Kc/sec at half-maximum. Without the mask on the lamp it is about 1.9 Kc/sec. These relatively narrow values make this device suitable for use as an accurate magnetic field measuring instrument. It has also been used successfully as part of a feed back loop to stabilize a magnetic field. In stronger fields an even greater advantage may be obtained by locking in the detector to one of the multiple Paschen–Back lines which has an even narrower width.

A more direct way to determine the nuclear momenta is as follows: The sample is subjected to a homogeneous magnetic field of a few gauss or more. The strong magnetic field decouples the I and J angular momenta, and a Paschen–Back effect takes place instead of the Zeeman hyperfine splitting. A study of this effect[17,18] shows that the number of resonances swept by the modulation field is $2I+1$. The line splitting is observed with the optical equipment placed in a set of Helmholtz coils 30 in. in diameter and at a field of 4 G. Homogeneity of the field is essential to observe the effect, and the field is modulated at a rate of 2 cps. This method was successfully tried in our laboratory.

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[17] G. Herzberg, *Atomic Spectra and Atomic Structure* (Dover Publications, New York, 1944), Chap. V.

[18] L. Anderson and A. Ramsey, Phys. Rev. **132**, 712 (1963).