

## Nuclear Spin and Magnetic Moment of 3.1-hr Cs<sup>134m</sup>†

L. S. GOODMAN AND S. WEXLER  
Argonne National Laboratory, Lemont, Illinois

(Received March 16, 1955)

The nuclear spin, hyperfine structure constant and magnetic moment of the excited isomer 3.1-hr Cs<sup>134m</sup> were determined by means of the atomic beam magnetic resonance method. The intensity of the active species was measured by deposition of the beam on a cold surface and subsequent counting. A spin of  $8(\hbar)$ , a hfs  $\Delta\nu = 3684.3 \pm 0.5$  Mc/sec, and a nuclear moment of  $1.10 \pm 0.01$  nm were found. The nuclear magnetic moment proved to be positive in sign.

### I. INTRODUCTION

IN recent years there has been a rising interest in the direct determination of nuclear spins and moments of radioactive isotopes. This interest has stemmed from advances in the understanding of nuclear structure as evidenced in such contributions as the shell model, beta-ray theory, and isomeric decay systematics.

One of the most fruitful methods of performing these measurements has been the atomic beam magnetic resonance method, developed for stable nuclides by Rabi and his co-workers.<sup>1</sup> A variation of the technique, introduced by Zacharias,<sup>2</sup> greatly increased the sensitivity of the method and led to the first atomic beam measurements on artificially produced active species.<sup>3</sup> The isotopic enrichment of the active nuclides was sufficiently high in these experiments that a conventional mass spectrometer could differentiate signals from adjacent masses despite the large amount of inactive material present. However, the enrichment of radioactive isotopes by neutron capture in chain reacting piles generally is not great enough to permit use of a mass spectrometer. In these cases measurement of the activity in the deposited beam may be resorted to, as was first demonstrated in the determination of the nuclear constants of Na<sup>24</sup> and K<sup>42</sup>.<sup>4</sup> All told, the nuclear properties of approximately twelve radioactive nuclides have been determined by atomic beam methods.<sup>2-6</sup> The active species selected for study have, up till now, been exclusively beta emitters, and the results obtained have accordingly served as tests of the predictions of the shell-model and beta-ray theories.

It was therefore desirable to measure the nuclear spin and magnetic moment of an excited isomer, since the results would be of interest with regard to the formulation of decay schemes based on semiempirical<sup>7</sup> and

theoretical<sup>8</sup> properties of gamma emission. The 3.1-hr nuclide Cs<sup>134m</sup> was chosen because (a) it could be prepared in sufficiently high specific activity by neutron capture that its beam intensity may be measured by condensation and subsequent counting, and (b) the excellent detector available for the inactive Cs<sup>133</sup> atoms in the beam made easy the calibration of the magnetic field. The well-known "flop-in" nuclear resonance experiment of Zacharias<sup>2</sup> was followed with an apparatus, which though similar in magnet geometry to an M.I.T. machine,<sup>3</sup> contained several modifications necessary for working with relatively short-lived activities.

### II. THEORY OF THE METHOD

Although the principles of the method have been described in detail elsewhere,<sup>1-3,9</sup> for the sake of clarity we will review its salient features. The experiment is based on the observation of transitions between magnetic substates in the Zeeman structure of the hyperfine levels of an atom in an external magnetic field. In the absence of an external field the angular momentum of the nucleus,  $I$  (in units of  $\hbar$ ), is coupled to that of the surrounding electrons, so that for an atom of electronic state  $J = \frac{1}{2}$  the total angular momentum of the atom,  $F$ , is either  $I + \frac{1}{2}$  or  $I - \frac{1}{2}$ . Since the electronic and nuclear spins may be coupled in two different ways, the magnetic moments associated with them will interact so that the energy of the atom will have either of two values, the difference between the states being the hyperfine structure separation  $\Delta W = \hbar\Delta\nu$ . Splitting of each of the hyperfine levels occurs when the atom is in an external magnetic field, as shown in Fig. 1, which describes some of the energy states of an atom with arbitrary nuclear spin  $I$  and  $J = \frac{1}{2}$ . Coupling of the nuclear and electronic spins is preserved in fields of low strengths, and the splitting is linear with field and equal between adjacent  $m_F$  sublevels. The difference in energy between the substates farthest apart in the  $F = I + \frac{1}{2}$  hyperfine level ( $m_F = F$  and  $m_F = -F$ ) is, neglecting the nuclear moment, just that due to the orientation of the electronic magnetic moment parallel and antiparallel to the external field, i.e.,  $2g_J J \mu_0 H$

† Work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> Rabi, Zacharias, Millman, and Kusch, Phys. Rev. **53**, 318 (1938); see also Kusch, Millman, and Rabi, Phys. Rev. **57**, 765 (1940), and later papers.

<sup>2</sup> J. R. Zacharias, Phys. Rev. **61**, 270 (1942).

<sup>3</sup> Davis, Nagle, and Zacharias, Phys. Rev. **76**, 1068 (1949).

<sup>4</sup> E. H. Bellamy and K. F. Smith, Phil. Mag. **44**, 33 (1953).

<sup>5</sup> Jaccarino, Bederson, and Stroke, Phys. Rev. **87**, 676 (1952).

<sup>6</sup> A. Lemonick and F. M. Pipkin, Phys. Rev. **95**, 1356 (1954).

<sup>7</sup> M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

<sup>8</sup> Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

<sup>9</sup> J. B. M. Kellogg and S. Millman, Revs. Modern Phys. **18**, 323 (1946).

(where  $g_J$  is the Landè  $g$ -factor,  $J$  the electronic angular momentum,  $\mu_0$  the Bohr magneton, and  $H$  the magnetic field). Since this energy is divided by  $2F+1$  equally separated levels into  $2F$  intervals, the Zeeman splitting of adjacent substates is  $g_J J \mu_0 H / F$ . For an  $S_{\frac{1}{2}}$  electronic state the frequency representing this energy difference is:

$$\nu_0 = \frac{\mu_0 H}{h F} = \frac{1.400 H}{I + \frac{1}{2}} \text{ Mc/sec.} \quad (1)$$

However, owing to the use of the Zacharias conditions<sup>2</sup> for refocusing the beam, only those transitions between substates are observed in which there is a reversal in sign of the effective electronic magnetic moment. Referring again to Fig. 1, we see that we are limited to the  $F = I + \frac{1}{2}$ ;  $m_F \leftrightarrow m_F = -(F-1)$  transition at low external fields and low frequencies. The assignment of the nuclear spin is a straightforward procedure, for it is only necessary to search for a single resonance corresponding to one of the various possible values of  $F = I + \frac{1}{2}$  in a known external magnetic field.

If, however, the nuclear spin of the isotope under study is large and/or the hfs rather small, the finite width of the radiofrequency resonance in this experiment may not permit resolution of the  $\nu_0$ 's for adjacent possible values of  $I$  at fields where the splitting is still linear. In this case the observation of a single resonance does not allow an unambiguous spin assignment. It is necessary to observe transitions in higher fields where the low frequency resonance deviates from Eq. (1). Under these conditions a better approximation for the  $F = I + \frac{1}{2}$ ;  $-F \leftrightarrow -(F-1)$  transition is<sup>3</sup>

$$\nu = 1.400 \frac{H}{F} + \left( 1.400 \frac{H}{F} \right)^2 \frac{2I}{\Delta\nu} \quad (2)$$

Measurement of the resonance frequencies at two settings of the external field will then permit calculation of the magnitude of the nuclear spin and an approximate value of the hfs. The external magnetic field is determined by observing the same transition in an atom of known nuclear constants, in our case, the inactive isotope Cs<sup>133</sup> present in the beam.

In increasingly higher external fields the decoupling of the nuclear and electronic spins becomes greater and the splitting of the sublevels departs from the description of Eq. (2). The behavior of the hyperfine structure in an external magnetic field is completely described for  $J = \frac{1}{2}$  by the Breit-Rabi equation,<sup>10</sup> which incidentally is the basis for the plot in Fig. 1 for arbitrary  $I$  and positive nuclear moment. After the nuclear spin has been assigned by the procedure indicated above, an accurate determination of hyperfine structure separation is carried out by observing the  $F = I + \frac{1}{2}$ ;  $m_F \leftrightarrow m_F = -(F-1)$  transition in successively larger fields until the Paschen-Back effect is strong. The hfs

<sup>10</sup> G. Breit and I. I. Rabi, Phys. Rev. **38**, 2082 (1931).

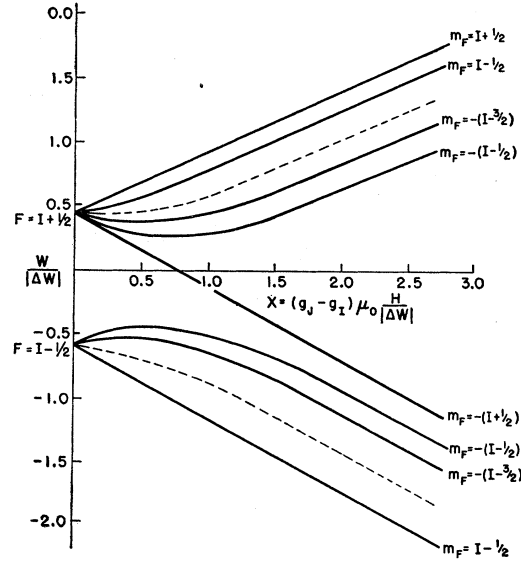


FIG. 1. Diagram showing variation with magnetic field of hyperfine substates of an atom with electronic angular momentum  $J = \frac{1}{2}$  and nuclear spin  $I$ . The nuclear moment is assumed positive. There are  $2(I + \frac{1}{2}) + 1$  substates in the  $F = I + \frac{1}{2}$  hyperfine level, of which five are shown, and  $2(I - \frac{1}{2}) + 1$  substates in the  $F = I - \frac{1}{2}$ , of which only three are drawn. The separation of adjacent sublevels has not been drawn to scale.

is then calculated for each of the frequencies  $\nu$  found by the use of the Breit-Rabi equation, which, for the low-frequency transition observed, may be put in the form<sup>11</sup>

$$\Delta\nu (\text{Mc/sec}) = \frac{(\nu - g_I k)(g_J k - \nu)}{\nu - g_I k - (g_J - g_I)k / (2I + 1)}, \quad (3)$$

where  $k = \mu_0 H / h$ , a constant of the external field, and  $g_I$  is the nuclear  $g$ -factor ( $= -\mu_I / I$ ). A value for  $g_I$  of Cs<sup>134m</sup>, sufficiently accurate for inclusion in Eq. (3), is obtainable from the Fermi-Segrè relation,<sup>12</sup>

$$\Delta\nu_{134m} / \Delta\nu_{133} = \frac{(2I_{134m} + 1)g_I^{134m}}{(2I_{133} + 1)g_I^{133}}, \quad (4)$$

where one takes the approximate  $\Delta\nu_{134m}$  derived in Eq. (2). Use is made of the known nuclear constants of the stable isotope Cs<sup>133</sup>:<sup>13</sup>  $\Delta\nu = 9192.76$  Mc/sec,  $I = 7/2(\hbar)$ ,  $\mu_I = 2.575$  nm. The more precise  $\Delta\nu_{134m}$ 's thus calculated yield better values of  $g_I^{134m}$  by introducing the former back into Eq. (4). From the improved  $g$ -factors the magnetic moment of Cs<sup>134m</sup> ( $\mu_I^{134m}$ ) is readily calculated in nuclear magnetons from  $\mu_I = -g_I I$ .

The sign of the nuclear moment may be decided upon from the calculation of the magnitude of the hfs. Taking  $g_I^{134m}$  first positive and then negative, we may compute from Eq. (3) two different values of  $\Delta\nu$  for each of the

<sup>11</sup> S. Millman and P. Kusch, Phys. Rev. **58**, 438 (1940).

<sup>12</sup> E. Fermi and E. Segrè, Z. Physik **82**, 729 (1933).

<sup>13</sup> H. Taub and P. Kusch, Phys. Rev. **75**, 1481 (1949); V. W. Cohen, Phys. Rev. **46**, 713 (1934); Kusch, Millman, and Rabi, Phys. Rev. **55**, 1176 (1939).

$\Delta F=0$  frequencies measured. One or the other set of such calculated hyperfine separations will be in agreement with the magnitude of  $\Delta\nu$  determined directly by observation of  $\Delta F=1$  transitions. Thus, comparison of the result from a high-frequency measurement with those calculated from low frequency  $\Delta F=0$  data resolves unambiguously the sign of  $gI^{134m}$ , and therefore of  $\mu I^{134m}$ .

When the two calculated sets of hfs differ by several Mc/sec, great precision in the direct transition measurement is unnecessary. The spectrum of  $\Delta F=1$  lines need only be observed in an external field sufficiently low that the Zeeman splitting is linear and the individual  $\Delta m_F = \pm 1$  transitions are unresolved. Under these conditions the frequency of a given  $\Delta F=1$  line in  $\text{Cs}^{134m}$  will be (see Rabi *et al.*<sup>1</sup>)

$$\nu = \Delta\nu + (2m_F' \pm 1) \frac{1.400H}{I + \frac{1}{2}}, \quad (5)$$

where  $m_F'$  can have any half-integral value between  $(I - \frac{1}{2})$  and  $-(I - \frac{1}{2})$ . Since  $m_F'$  varies only in jumps of one, the spacing between adjacent lines in the unresolved pattern is constant. With the exception of the  $F = I + \frac{1}{2}$ ,  $m_F = -(I + \frac{1}{2}) \leftrightarrow F = I - \frac{1}{2}$ ,  $m_F = -(I - \frac{1}{2})$  transition, which is not observed in this experiment, and the  $F = I + \frac{1}{2}$ ,  $m_F = (I + \frac{1}{2}) \leftrightarrow F = I - \frac{1}{2}$ ,  $m_F = (I - \frac{1}{2})$ , each line is the superposition of two transitions. Thus if the number of possible  $\Delta F=1$  transitions is large,

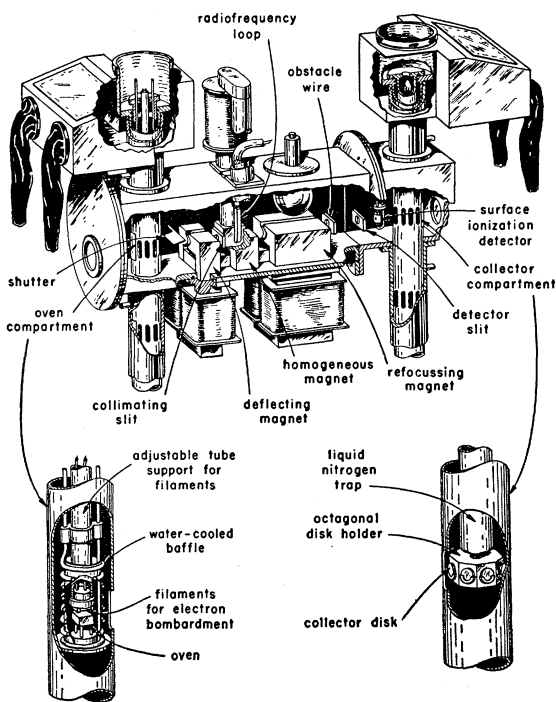


FIG. 2. Artist's sketch of atomic beam apparatus. Only the important components are shown. Many of the details, such as the pumping systems, inside section walls and hydraulic lifts have been left out to avoid confusion.

i.e.  $I$  is high, the unresolved pattern should be symmetrical in shape, the center being very close to the frequency of the hyperfine structure constant.

An estimate may be made of the width of the unresolved pattern. From Eq. (5) neglecting the finite resolution of the apparatus, we find the frequency difference between extreme lines is  $4I(1.400H)/(I + \frac{1}{2})$  or  $4I\nu_0$ . If the external magnetic field is such that the Zeeman splitting is 50 kc/sec, the width will be approximately 1.6 Mc/sec when  $I$  is taken as  $8(\hbar)$ . The actual width, however, would be somewhat greater because of the short time of the atom in the rf field and because of the adding up of tails from adjacent resonances.

### III. APPARATUS

#### A. General Plan of Apparatus

The atomic beam magnetic resonance apparatus constructed for this experiment was patterned after the M.I.T. machine described by Davis, Nagle, and Zacharias.<sup>3</sup> A cut-away artist's sketch appears in Fig. 2. The usual inhomogeneous deflecting and refocusing fields, with gradients parallel and in the same direction, are on either side of the homogeneous field in which transitions between the magnetic sublevels take place. Atoms emerging from the oven pass from the oven compartment through an isolation chamber into the main section housing the magnets and detectors. Depending on the velocities of the atoms, the angles from the normal at which they leave the oven, and the gradient of the deflecting field, certain ones will be focused on the collimating slit and pass into the gap containing the homogeneous and oscillating radio-frequency fields. Only those atoms which in the overlapping fields undergo transitions involving a change in sign of their effective magnetic moments at high field are refocused onto the detector slit, lying, like the oven and collimating slits, on the neutral axis of the beam. Atoms not suffering such transitions are deflected away from the detector by the two inhomogeneous magnets.

Radiofrequency power to effect the transitions was introduced through a vertical copper "hairpin," two cm along the beam, situated between the poles of the uniform field magnet. The oscillating field produced was mainly parallel to the beam and at right angles to the static field. A "pickup" coil mounted in the vicinity of the hairpin served to give a measure of the power of the oscillating field. The rf signal picked up by the coil was rectified by a crystal, and the resultant current measured with a microammeter. For the low-frequency studies a Kovar seal made a satisfactory vacuum lead through for the rf. In the measurements of the  $\Delta F=1$  transitions, which occurred at relatively high frequencies, an Andrews Corporation gas barrier seal, matched to  $\frac{7}{8}$ -inch air dielectric coaxial line, was used, and was found to be negligibly lossy while being quite satisfactory vacuum-wise.

The widths of all the slits defining the beam—oven, collimating and detector—were each 0.010 inch. The obstacle wire measured 0.015 inch.

A conventional type of surface ionization detector, mounted through a flange in the side of the apparatus and situated directly behind the detector slit, served to measure the Cs<sup>133</sup> intensity in the beam. The detector rode on a carriage which could be moved out of the path of the beam without disturbing the vacuum. Positive ion currents to the cylindrical collector plate were amplified by a vibrating reed electrometer.

The active Cs<sup>134m</sup> component was measured by condensing the beam on one-inch diameter copper disks ( $\frac{1}{16}$ -inch thick), which were held by spring clamps on the faces of a brass octagon attached to the bottom of a liquid nitrogen trap. These disks were washed with water, alcohol, and ether before being used. If a disk showed radioactive contamination it was first etched in dilute HCl. No other surface treatment was necessary. The long tube containing the collector was rotated in o-ring gaskets so as to position each of eight disks in turn behind the collector slit. Vertical movement of the tube was effected by hydraulic lifts. A drybox attached around the tube above the apparatus made it possible to bring the cold octagon and trap through vacuum locks into a dry helium atmosphere for the changing of disks. Measurement of the activity was made by placing each disk in a windowless G-M flow counter containing a 98 percent He-2 percent Isobutane gas mixture.

### B. Vacuum System

The vacuum housing consisted of a brass box 12 in. wide,  $13\frac{3}{4}$  in. high, and 55 in. long, supported by an aluminum Unistrut frame. Walls of the box,  $\frac{3}{4}$  in. thick on sides and top and 1 in. on bottom, were either welded together and then tinned on the inside or screwed together and silver soldered. Components inside the housing were introduced through appropriately cut holes in the four sides and held on by flanges bolted to the apparatus. o-ring gaskets, greased with Apiezon T, formed the vacuum seals.

Evacuation of the apparatus was performed by two 6-in. oil diffusion pumps attached to the main chamber, and one 4-in. pump each on the isolation and oven compartments. The four pumps were backed by a 4-in. booster pump which evacuated into a mechanical pump. Each of the four primary pumps was attached to the side of the vacuum housing through a water-cooled 4-in. baffle valve and a large, cylindrical liquid nitrogen trap. An auxiliary system consisting of 2-in. tubing and valves evacuated by a large mechanical pump served the dual purposes of roughing down the chambers and tubes and of maintaining a vacuum between o-ring pairs in which shafts leading into the apparatus were rotated. A vacuum of less than  $10^{-6}$  mm

pressure was readily attained in the main and isolation chambers, while the pressure in the oven section was usually between  $10^{-6}$  and  $10^{-5}$  mm.

### C. Magnets

Dimensions of the pole pieces of the two deflecting magnets were identical with those designed by Nagle<sup>14</sup>; however, the gap of the homogeneous field was increased to  $\frac{1}{2}$  in. in our apparatus. The pole pieces of all three magnets were bolted onto a brass bar, which served to maintain alignment. The magnetic circuits were brought out through the vacuum wall via flanged o-ring seals and were energized by high-resistance coils outside the housing. This scheme allowed the use of electronically regulated current supplies and obviated the need for large storage batteries and charging equipment.

### D. Oven

A cold-rolled steel oven of simple design (Fig. 3) sat on a tripod in the oven tube (Fig. 2), two of whose legs constituted a tantalum-tungsten thermocouple, with the oven forming the junction. This scheme allows monitoring of oven temperatures from 100 to over 1500°C. The tripod was mounted on a carriage installed on a base in the long oven tube. Controls for fine adjustment (such as direction, tilt and traverse position of the oven slit) entered through the top flange of the tube and were connected to the carriage by means of flexible rods.

The heating system was designed to be relatively universal; electron bombardment from thoriated tungsten filaments was decided upon. For Cs metal, of course, just the radiation from the filaments give adequate heating. The heating and temperature monitoring designs had the added advantage of not requiring connections to the oven block itself, which contained a radioactive source. Introducing the active Cs metal into the oven was done in a He-filled drybox attached to the oven tube above the apparatus. Like the collector, this tube was moved vertically through vacuum locks by hydraulic lifts.

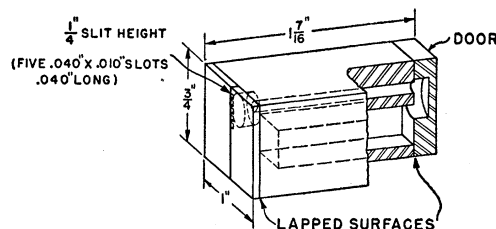


FIG. 3. Cold-rolled steel oven for Cs metal. Two quartz capsules are placed in the cavity and then partially crushed. The lapped door and slit jaws are held onto the block by screws not indicated in the drawing.

<sup>14</sup> D. E. Nagle, thesis, Massachusetts Institute of Technology, 1948 (unpublished).

### E. Radio-Frequency Equipment

For study of the low-frequency  $\Delta F=0$  transitions, the General Radio Type 805-C signal generator (50 kc/sec to 50 Mc/sec) was used. A conventional, one stage, tuned power amplifier, with a 2E26 tube, was used to enhance the transition probability in this frequency region. In the region extending up to 300 Mc/sec a Hewlett-Packard Model 608 A was employed; a Spencer-Kennedy Model 202 wide band amplifier used with this oscillator gave some improvement in the refocused beam intensity.  $\Delta F=1$  transitions were excited by a Sperry reflex klystron, Model 2K-42, powered by a Polytechnic Research and Development Company Type 801 A supply.

Frequencies less than 150 Mc/sec were measured by a Berkeley Frequency Meter, Model 5570, with a range extender, Model 5575. The instrument is capable of accuracies of one part in  $10^7$ . In the present work, however, no effort was made to measure frequencies to better than 1 kc/sec. For measurement of frequencies higher than 150 Mc/sec, the signal from the H-P 608 A was crystal mixed with that from the G-R 805 C, the output frequency of which was determined by the Berkeley meter. The 600-kc/sec beat was detected with a NC-125 receiver, which was calibrated at that point. The same technique worked well in the measurement of the klystron frequency, in which case the Hewlett-Packard 608 A served as the local oscillator and a Hallicrafters Model S-36 A as receiver for the 33-Mc/sec beat between the 25th harmonic of the H-P and the signal from the klystron. To decide whether the klystron frequency was higher or lower than that of the local oscillator harmonic, the signal from the pickup loop was measured to within 15 Mc/sec by means of a crystal rectifier and variable tuning stub arrangement. Drift of the H-P 608 A (about 1 kc/sec) and fluctuation of the power level of the klystron allowed about 40 kc/sec uncertainty in the klystron frequency.

### IV. PRELIMINARY ALIGNMENT AND PROCEDURE

After optical alignment of the slits with a telescope, a sodium beam was brought up, deflected with the inhomogeneous magnets, and refocused by applying a resonant combination of uniform field and rf frequency. Strong stray fields from the inhomogeneous magnets induced a field of about 100 gauss in the gap of the uniform field magnet. This field was "bucked out"

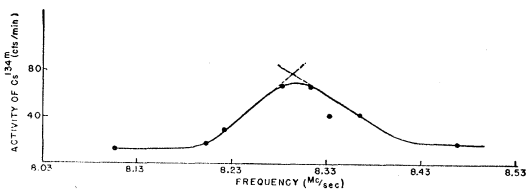


Fig. 4. Typical resonance curve observed for  $F=I+\frac{1}{2}$ ;  $m_F = -F \rightarrow m_F = -(F-1)$  transitions.

when necessary by reversing the current in the magnet winding of the homogeneous field. The magnitudes of the two deflecting fields and the positions of the collector and oven slits were varied over wide ranges so as to obtain the maximum signal to background ratio. This ratio was thereby improved by a factor of two to three over that found with the arrangement suggested by optical alignment alone.

Cesium metal, distilled from a mixture of Ca chips and CsCl, was sealed under vacuum in small quartz vials; each capsule contained about 350 mg of material. The 3.1-hr  $\text{Cs}^{134m}$  isomer was produced by irradiation for several hours in a thermal neutron flux of about  $1.5 \times 10^{13}/\text{sec}/\text{cm}^2$  available in the heavy water reactor at this laboratory. In a given run two such irradiated vials were partially crushed in the oven, situated in the helium atmosphere of the drybox, and the closed oven was placed on the tripod in the raised oven tube. This loading usually lasted over seven hours. The tube was then lowered through vacuum locks so as to bring the oven into position. After the cesium beam was stabilized by adjusting the oven temperature, the obstacle wire was moved into position and the deflecting

TABLE I. Observed transition frequencies for  $\text{Cs}^{134m}$  and  $\text{Cs}^{133}$  and calculated hfs  $\Delta\nu_{134m}$  (Mc/sec).

$\nu_{133}$	$\nu_{134m}$	$\Delta\nu_{134m}$ if $\mu I$ pos	$\Delta\nu_{134m}$ if $\mu I$ neg
$9.365 \pm 0.010$	$4.480 \pm 0.015$		
$17.220 \pm 0.010$	$8.295 \pm 0.020$		
$43.940 \pm 0.010$	$21.934 \pm 0.020$		
$90.937 \pm 0.010$	$48.403 \pm 0.010$	$3681 \pm 5$	$3698 \pm 5$
$164.105 \pm 0.030$	$97.140 \pm 0.025$	$3682 \pm 1$	$3689 \pm 1$
$282.845 \pm 0.020$	$201.666 \pm 0.020$	$3684.3 \pm 0.5$	$3695.0 \pm 0.5$

magnets were energized. The homogeneous field was set by observing the low-frequency  $\Delta F=0$  line in  $\text{Cs}^{133}$ , using the hot-wire detector. The collecting disks were lowered through vacuum locks, cooled to liquid nitrogen temperature, and brought into position.

Search for the  $\Delta F=0$  transition in active  $\text{Cs}^{134m}$  was then made by varying the rf, usually in 50–100 kc/sec steps, and exposing one of the disks to the beam for a definite time (usually five minutes) with each frequency setting. Recalibration of the homogeneous field was conducted between deposition periods. After collection of the beam on all eight disks, the collector tube was raised into the drybox and the disks were removed for counting. The 100-keV conversion electron activity on each was measured in a windowless flow type G-M counter. The disks were each counted at least twice and the decay rates at a common time determined for plotting of a resonance curve. On the following day each disk was again counted to be sure of the absence of possible 2-yr  $\text{Cs}^{134}$  contamination from handling the collectors. When a resonance was found, the search of the frequency region was repeated. The observation of a given  $\Delta F=0$  transition in  $\text{Cs}^{134m}$  provided values of

$\Delta\nu_{134m}$  and  $g_I^{134m}$  from which the frequency region to be searched at higher external field could be calculated.

In the direct determination of the hyperfine structure constant the homogeneous magnetic field was set to produce a Zeeman splitting in Cs<sup>134m</sup> of approximately 50 kc/sec. A search was then conducted for a broad refocused intensity in the frequency regions suggested by the two  $\Delta\nu$ 's calculated from the low-frequency data. The reflex klystron was varied in 500-kc/sec steps, and its output kept constant to within 40 kc/sec for the duration of each beam collection period.

## V. CALCULATIONS AND RESULTS

The measured frequencies of the  $F=I+\frac{1}{2}$ ;  $m_F = -F \leftrightarrow m_F = -(F-1)$  transitions in the Cs<sup>134m</sup> isomer are listed in Table I with the corresponding lines of Cs<sup>133</sup> used to measure the magnitude of the external field. Observations were first made in sufficiently low magnetic field to preserve largely the coupling of the electronic and nuclear spins, and then extended gradually to higher fields in which the decoupling of the angular momenta and the very small direct interaction of the nuclear magnetic moment with the applied field measurably affect the magnitude of the Zeeman splitting. From the theoretical discussion it is clear that we are interested in extracting the following information about 3.1-hr Cs<sup>134m</sup> from the data: the nuclear spin  $I$  (in units of  $\hbar$ ), the hyperfine structure separation (in Mc/sec), the nuclear magnetic moment (in nuclear magnetons), and the sign of the moment.

The two lowest frequency resonances (a typical curve appears in Fig. 4) observed in Cs<sup>134m</sup>, 4.480 and 8.295 Mc/sec (Table I), were introduced in Eq. (2) to evaluate the nuclear spin and to give an approximate value for the hyperfine structure constant. A nuclear angular momentum of  $8(\hbar)$  and a  $\Delta\nu \approx 3600$  Mc/sec come out of the calculation. These values were confirmed by the measurements at higher frequencies. From the approximate  $\Delta\nu$  so found and the Fermi-Segrè relation [Eq. (4)], a sufficiently accurate  $g_I$  was evaluated for use in the Breit-Rabi equation [Eq. (3)]. More accurate hyperfine structure constants were then calculated for each of the resonances found at progressively higher rf signals. The frequencies of the observed transitions are assembled in Columns 1 and 2 of Table I along with their estimated errors, these assumed to be due only to the uncertainty in estimating the exact position of the resonance. As pointed out under *Theory of Method*, either of two values for  $\Delta\nu$  is the correct one depending on whether the nuclear magnetic moment is parallel or antiparallel to the spin. Thus the correct hfs is most nearly  $3684.3 \pm 0.5$  Mc/sec (Column 3, Table I) if the moment is positive and  $3695.0 \pm 0.5$  Mc/sec (Column 4) if negative. Although the consistency of the  $\Delta\nu$  values so calculated can sometimes<sup>1-3</sup> indicate which of the two signs is to be taken, this was not the case here, and only the

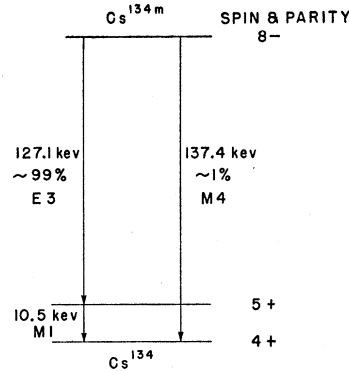


FIG. 5. Decay systematics of Cs<sup>134m</sup> [Sunyar, Mihelich, and Goldhaber (see reference 17)].

measurement of the direct hyperfine transition in Cs<sup>134m</sup> could settle the question. Because of the conveniently low frequency of the hfs,  $\Delta F=1$  transitions could be looked for with available equipment. Operating in an external magnetic field so low as to permit observation only of the unresolved lines, we found a broad resonance, about 2.5 Mc/sec wide at half-maximum, centered at  $3684.5 \pm 0.5$  Mc/sec, while no indication of a resonance was seen in the neighborhood of 3695 Mc/sec. Therefore, we conclude that the hyperfine structure constant of 3.1-hr Cs<sup>134m</sup> is  $3684.5 \pm 0.5$  Mc/sec and the nuclear magnetic moment is positive. Finally, from the Fermi-Segrè formula  $\mu_I^{134m} = +1.10 \pm 0.01$  nm.

Substantially the same values for the nuclear constants of Cs<sup>134m</sup> reported here and in preliminary notes<sup>15</sup> were obtained by Cohen and Gilbert<sup>16</sup> in a similar, but independent experiment at the Brookhaven National Laboratory.

## VI. DISCUSSION

For some time it had been assumed that 3.1-hr Cs<sup>134m</sup> decayed by emission of a single  $\gamma$  ray of 127-keV (an  $E3$  transition) to a 2.3-yr Cs<sup>134</sup> ground state. However, as a consequence of the preliminary measurements on the spin of the excited isomer, Sunyar, Mihelich, and Goldhaber<sup>17</sup> (Fig. 5) reinvestigated this decay and found a previously undetected 10.5-keV gamma following the 127-keV transition. These authors presented evidence for the decay systematics of the Cs<sup>134m</sup> isomer presented in Fig. 4. From conversion coefficient data, and empirical and theoretical considerations, the 127.1-keV  $\gamma$  ray was considered to be an  $E3$  transition, the 10.5 keV an  $M1$  decay, and the 137.4-keV cross over an  $M4$ . As the spin of the 2.3-yr state has been found to be  $4(\hbar)$  by direct measurement using atomic beam techniques,<sup>4,5</sup> the decay systematics of the Cs<sup>134m</sup> transitions favor a

<sup>15</sup> L. S. Goodman and S. Wexler, Phys. Rev. **95**, 570 (1954); **97**, 242 (1955).

<sup>16</sup> V. W. Cohen and D. A. Gilbert, Phys. Rev. **95**, 569 (1954); **97**, 243 (1955).

<sup>17</sup> Sunyar, Mihelich, and Goldhaber, Phys. Rev. **95**, 570 (1954).

spin of  $8(\hbar)$  for the 3.1-hr isomer. Since this result agrees with the directly measured value, the present experiment serves as evidence for the validity of decay schemes based on calculations of internal conversion coefficients by Rose *et al.*<sup>3,18</sup>

From the shell model two possible proton-neutron configurations are possible for the excited isomer  $\text{Cs}^{134m}$  compatible with the measured spin of  $8(\hbar)$ . These are<sup>19</sup>  $1g_{7/2}, 1h_{11/2}$ , and  $2d_{5/2}, 1h_{11/2}$ . Using a two-nucleon wave function, one arrives at the expression of Bellamy and Smith<sup>4</sup> for the magnetic moments of odd-odd nuclei, from which the moments associated with these states are calculated to be  $-0.36$  and  $+2.90$  nm, respectively. Possibly a mixed configuration, as suggested by de-Shalit and Goldhaber,<sup>20</sup> of these two states may account for the measured result of  $+1.10$  nm.<sup>21</sup>

An empirical method for estimating moments of

<sup>18</sup> Rose, Goertzel, and Swift (privately circulated tables).

<sup>19</sup> Mayer, Moszkowski, and Nordheim, *Revs. Modern Phys.* **23**, 315 (1951).

<sup>20</sup> A. de-Shalit and M. Goldhaber, *Phys. Rev.* **92**, 1211 (1953).

<sup>21</sup> The suggestion of a mixed configuration in  $\text{Cs}^{134m}$  was first made by Cohen and Gilbert, reference 15.

odd-odd nuclei stems from proposals<sup>22</sup> that the spin contributions to the magnetic moment of the odd nucleons are suppressed from their respective free particle values when the neutron and proton are in the nucleus. Following this suggestion, we may take the effective intrinsic moment of the odd neutron and odd proton in  $\text{Cs}^{134m}$  to be  $-1.4$  and  $1.5$  nm, respectively, using the graph prepared by Bloch.<sup>22</sup> Introducing these values into the same two-nucleon calculations, we find fair agreement with experiment in the result of  $+0.8$  nm computed for the  $g_{7/2}, h_{11/2}$  configuration. The  $d_{5/2}, h_{11/2}$  proton-neutron configuration gives the much higher value of  $+2.1$  nm for the nuclear moment.

#### ACKNOWLEDGMENTS

We wish to thank J. A. Dalman for his ingenious design work and assistance in performing the experiments, H. W. Ostrander for his design of many parts of the apparatus, and M. Hamermesh, D. E. Nagle, and D. Kurath for stimulating discussions.

<sup>22</sup> F. Bloch, *Phys. Rev.* **83**, 839 (1951); H. Miyazawa, *Progr. Theoret. Phys. (Japan)* **6**, 263 (1951); A. de-Shalit, *Helv. Phys. Acta* **24**, 296 (1951).

## Cosmic-Ray Intensity above the Atmosphere at High Latitudes\*†

L. H. MEREDITH,‡ J. A. VAN ALLEN, AND M. B. GOTTLIEB  
*Department of Physics, State University of Iowa, Iowa City, Iowa*

(Received October 29, 1954; revised manuscript received April 8, 1955)

The total charged particle cosmic-ray intensity above the atmosphere has been measured with thin-walled Geiger counters (total effective stopping power of apparatus and residual atmosphere  $0.5$  g/cm<sup>2</sup> of aluminum) carried in balloon-launched rockets at geomagnetic latitudes  $54.3^\circ$  N,  $62.1^\circ$  N,  $71.9^\circ$  N, and  $86.7^\circ$  N. The respective values of unidirectional particle intensity averaged over the upper hemisphere are:  $J = 0.44 \pm 0.01$ ,  $\leq 0.50 \pm 0.05$ ,  $\leq 0.50 \pm 0.05$ , and  $= 0.48 \pm 0.01$  (cm<sup>2</sup> sec sterad)<sup>-1</sup>. These results are consistent with the complete or nearly complete absence of primary cosmic rays having a magnetic rigidity less than  $1.7 \times 10^9$  volts.

### I. INTRODUCTION

#### A. Purpose

THE purpose of this investigation was to make an absolute measurement of the total intensity of charged primary cosmic rays down to the lowest feasible value of magnetic rigidity  $R$ —in this case down to a value of  $R$  of about  $0.2 \times 10^9$  volts.

The intensity of low-rigidity cosmic rays is of interest for several reasons, *viz.*: (a) They may constitute a significant portion of the primary beam. (b) The presence or absence of low-rigidity primaries may

provide a crucial test of any proposed mechanism for the astrophysical origin and propagation of cosmic rays. (c) A specific aspect of (b) may be a definitive test of the magnitude of the magnetic dipole moment of the sun. (d) It is of aeromedical interest to know the radiation intensity which will be encountered in flight at high altitudes and in the vicinity of the earth.

#### B. Necessary Geographic Location

The theoretical properties of the geomagnetic field as a magnetic spectrometer are summarized by Alpher.<sup>1</sup> A special feature of the theory is that the critical value of magnetic rigidity  $R_c$ , which a particle must possess in order to reach the top of the atmosphere at a specified location, diminishes with increasing geomagnetic lati-

\* Based on the doctoral dissertation of L. H. M. (June, 1954).

† Assisted by joint program of the Office of Naval Research, the U. S. Atomic Energy Commission, and the Navy Bureau of Aeronautics; and by the Research Corporation.

‡ Present address: U. S. Naval Research Laboratory, Washington 25, D. C.

<sup>1</sup> R. A. Alpher, *J. Geophys. Research* **55**, 437 (1950).