This gives

$$a=(\beta+\alpha\delta^2)/(1+\delta^2)+O[(\beta-\alpha)^3], \tag{59a}$$

$$b=\beta-\tfrac{1}{2}(\beta-\alpha)/(1+\delta^2)+O[(\beta-\alpha)^3], \tag{59b}$$

$$s^2=1+\delta^2+O[(\beta-\alpha)^2]. \tag{59c}$$

For the coefficients A, B, and C we find in this case (again taking $u_0=v_0=0$).

(1) $w=u/M_0$

$$\left.\begin{aligned} A&=\frac{\delta}{1+\delta^2}\left(m_0-\frac{1+\delta^2}{\delta^2+\beta/\alpha}\right)+O(\beta^2),\\ B&=-\frac{m_0\delta}{1+\delta^2}+O(\beta^2),\\ C&=\frac{\delta}{1+\delta^2}\left[\frac{m_0(\beta-\alpha)(\frac{1}{2}-\delta^2)}{1+\delta^2}-\alpha\right]+O(\beta^3),\end{aligned}\right\} \tag{60}$$

(2) $w=v/M_0$

$$\left.\begin{aligned} A&=-\frac{\beta-\alpha}{(1+\delta^2)^2}\cdot\delta^2\left(m_0-\frac{1+\delta^2}{\delta^2+\beta/\alpha}\right)+O(\beta^3),\\ B&=\frac{1}{1+\delta^2}\left[\frac{(\beta-\alpha)\delta^2m_0}{1+\delta^2}+\alpha\right]+O(\beta^3),\\ C&=-m_0+O(\beta^2).\end{aligned}\right\} \tag{61}$$

(3) $w=M_z/M_0$

$$\left.\begin{aligned} A&=\frac{\delta^2}{1+\delta^2}\left(m_0-\frac{1+\delta^2}{\delta^2+\beta/\alpha}\right)+O(\beta^2),\\ B&=m_0/(1+\delta^2)+O(\beta^2),\\ C&=\alpha+m_0(\beta-\alpha)(\tfrac{1}{2}+2\delta^2)/(1+\delta^2)+O(\beta^3).\end{aligned}\right\} \tag{62}$$

PHYSICAL REVIEW VOLUME 76, NUMBER 8 OCTOBER 15, 1949

Atomic Beam Magnetic Resonance Experiments with Radioactive Elements Na^{22}, K^{40}, Cs^{135}, and Cs^{137}*

LUTHER DAVIS, JR., DARRAGH E. NAGLE, AND JERROLD R. ZACHARIAS
*Department of Physics and Research Laboratory of Electronics,** Massachusetts Institute of Technology, Cambridge, Massachusetts*

(Received July 12, 1949)

Modifications of the atomic beam magnetic resonance method for the determination of nuclear spins and moments by observation of the hyperfine structure of atomic ground states are described which make it possible to work with very small quantities in samples with low concentrations. These modifications include the analysis of the beam by means of a mass spectrometer while performing the resonance experiment. A new source of beams of atomic alkali metals is described. The results are shown in the following tabulation:

	Spin	h.f.s. $\Delta\nu$ mgc/sec.	Nuclear magnetic moment nuclear magnetons
Na^{22}	3	1220.64±0.04	1.746±0.003
K^{40}	4	1285.73±0.05	−1.290±0.005
Cs^{135}	7/2	9724 ±8	2.724±0.010
Cs^{137}	7/2	10,126 ±7	2.837±0.010

ATTEMPTS to compare the various forms of the Fermi theory of beta-ray disintegration with experiments usually meet with the difficulty that those characteristics of the decay which make it possible to observe the nuclear angular momentum directly are just those characteristics which make the observation of the energetics of the decay more difficult and less precise. In particular, the long half-lives of C^{14}, K^{40}, and Rb^{87} permit the use of large amounts of material in observation of nuclear spin,[1,2] but the determination of the shapes [3,4] of their beta-ray spectra, especially in the low energy regions, is less reliable than could be obtained with more active materials because of the effect of scattering in the source. For tritium, on the other hand, one can work with extremely active sources because the radiation (beta-rays of less than 18 kev) is all absorbed by the containing vessel, but only recently has the spectrum shape been observed.[5] It is therefore desirable to develop a method for the observation of nuclear moments which can be applied to more energetic substances with shorter half-lives, since it is for such materials that decay schemes and spectra can be well studied. Preliminary calculations and experience with K^{40} indicated that the molecular beam magnetic resonance method might well be applicable to radio isotopes of the alkali metals because of the excellent existing method of detection. The techniques described in the present paper, devoted to Na^{22}, K^{40}, Cs^{135}, and Cs^{137}, can be extended to other nuclear species with some additional development. From the point of view of radioactive decay, Na^{22} has been most carefully studied by Good, Peaslee, and Deutsch[6] and Cs^{137} is being extensively examined by a number of workers including Townsend, Cleland, and Hughes[7] and Mitchell and Peacock.[8]

---

* Preliminary reports of some of these results are given by Luther Davis, Jr., Phys. Rev. **74**, 1193 (1948), Phys. Rev. **76**, 435 (1949), and D. E. Nagle, Phys. Rev. **76**, 847 (1949).

** This work was supported in part by the Signal Corps, the Air Materiel Command, and the ONR.

[1] F. A. Jenkins, Phys. Rev. **74**, 355 (1948).

[2] J. R. Zacharias, Phys. Rev. **61**, 270 (1942).

[3] D. E. Alburger, Phys. Rev. **75**, 1442 (1949).

[4] C. S. Wu and R. D. Albert, Phys. Rev. **75**, 315 (1949).

[5] G. C. Hanna and B. Pontecorvo, Phys. Rev. **75**, 983 (1949); Curran, Angus, and Cockcroft, Phil. Mag. **40**, 53 (1949).

[6] Good, Peaslee, and Deutsch, Phys. Rev. **69**, 313 (1946).

[7] Townsend, Cleland, and Hughes, Phys. Rev. **74**, 499 (1948).

[8] A. C. G. Mitchell and G. L. Peacock, Phys. Rev. **75**, 197 (1949).

In considering various possible methods for the observation of nuclear moments one is struck by the requirement imposed by chemical manipulation of minute quantities of radioactive material. One is usually forced to considerable dilution and most of the possible methods are not at their best when working with great dilutions, especially with isotopes of the element under investigation. In the method of microwave spectroscopy, collision broadening by foreign atoms reduces the line intensity in proportion to the dilution. Band spectroscopic determination of nuclear spin requires a molecule with two similar atoms and this always sets a requirement of high concentration. Hyperfine structure measurement in the visible or near visible region of atomic spectra is likely to suffer only from overlapping due to strong lines and seems to be very promising for atoms with large isotope shift. In principle, the method to be described here can be used with such small amounts of material and with such great dilution that extension of it to rare isotopes with half-lives only long enough to permit manipulation in the apparatus seems possible.

THEORY OF THE METHOD

The experimental method is a modification of the well-known molecular beam magnetic resonance method which follows in many details the experiment described by Kusch, Millman, and Rabi[9] and that described by Zacharias[2] in a determination of the nuclear spin and magnetic moment of K^{40}. The power of the method depends on the ability to observe the Zeeman effect of the atomic ground state of the isotope in question in an externally applied magnetic field which is sufficiently weak to preserve strong coupling between the nuclear spin and the resultant angular momentum of the atomic electrons. In the case of the alkali atoms discussed in the present paper, the atomic ground state is $^2S_{\frac{1}{2}}$ and in weak field this combines with a nuclear angular momentum, I, to give resultants of $F=I+\frac{1}{2}$ and $F=I-\frac{1}{2}$. The preservation of this coupling can most easily be described by saying that the nucleus finds itself in a large magnetic field (of the order of 10^6 gauss) produced by the atomic electrons which are precessing about the direction of the applied field. The combined system then has the total angular momentum $F=I+J$ where J is the angular momentum of the atomic electron configuration. The magnetic moment, however, associated with this total angular momentum F is overwhelmingly that of the electrons. Thus the magnitude of the Zeeman splitting of the atomic ground state in weak field is very closely $eH/4\pi mcF$ or 1.400 H/F Mc/sec. Observation of the frequency of transition between two adjacent levels separated only by the Zeeman splitting in weak field yields a value of F and correspondingly a value of I. To determine I for a nucleus of unknown spin, one can successively assume integral values 0, 1, 2, 3, 4, 5, 6, 7, as possible spin values for nuclei with even mass number or 1/2, 3/2, 5/2, 7/2, 9/2, 11/2, or 13/2 for nuclei with odd mass number. In a known small magnetic field the search for radiofrequency transitions is extremely simple since it is only necessary to try to observe transitions at the frequencies 1.400 H/F Mc/sec. for the eight or nine assumed I values. It should be pointed out that no prior knowledge of the nuclear magnetic moment or gyromagnetic ratio is necessary or even useful in this search. The value of the small applied magnetic field can be easily determined beforehand by observing the same type of transition with an atom of known nuclear and atomic properties. This method can be applied to any atom with a known value of the electronic angular momentum provided it is not zero. It is unfortunately not applicable to such important elements as helium, beryllium, carbon, magnesium, etc. There is an additional complication present in samples containing the radio-isotope under investigation mixed with a large amount of other isotopes. If, for instance, both isotopes have the same spin then the Zeeman effect in weak field will occur at the same frequency. Obviously this is not possible with Na^{22} and Na^{23} because the former must have integral spin and the latter has a spin of 3/2. With the cesium isotopes of odd mass number, we find that the spins are all equal. There are two methods for avoiding this trouble, and fortunately they can be used simultaneously. The first of these depends on the fact that as the applied magnetic field is increased from weak values to strong

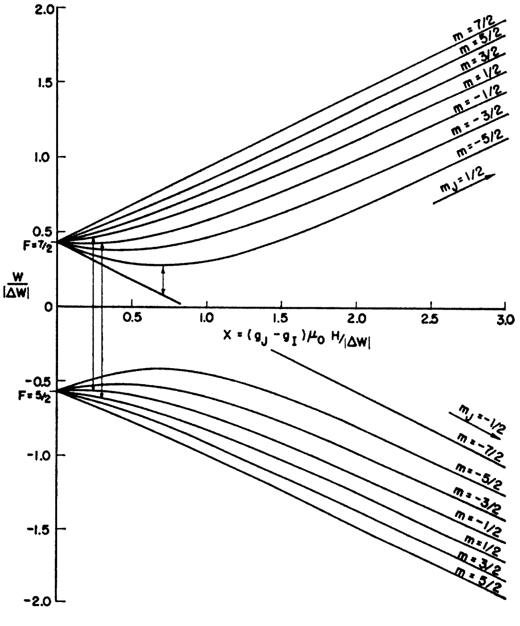

FIG. 1. Diagram to show the variation with magnetic field of the ground state energy levels for an atom with electronic angular momentum $J=\frac{1}{2}$ and nuclear angular momentum $I=3$. The nuclear magnetic moment is taken as positive. If the nuclear magnetic moment is negative the diagram is inverted and all m values change sign. The transitions marked by arrows are those observed in the present experiment with Na^{22}. The spin value is determined from observation of the transitions marked by the shortest of the three arrows.

---

[9] Kusch, Millman, and Rabi, Phys. Rev. **57**, 765 (1940).

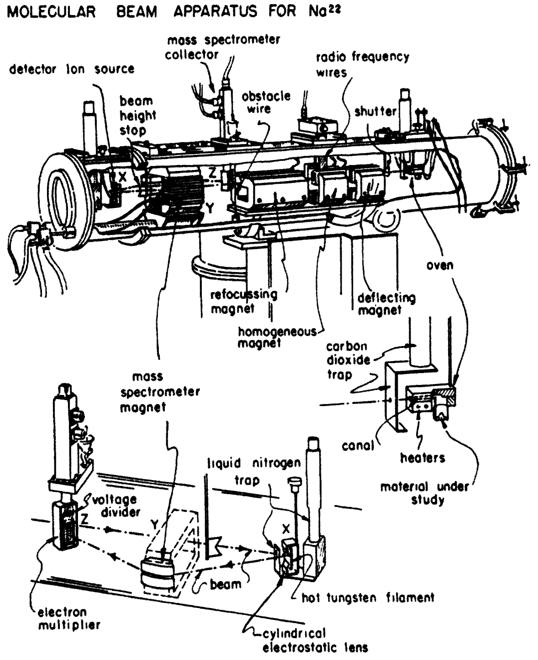

FIG. 2. Cut-away sketch of atomic beam apparatus indicating its important components. To avoid confusion, many details such as differential pumping chamber, pumping system, defining slots, and some magnet coils have been omitted.

values the coupling between I and J begins to break and the Zeeman splitting begins to depart from the value of $1.400\ H/F$. The behavior in the region intermediate between the Zeeman and the Paschen-Back region is completely described for $J=\frac{1}{2}$ by the Breit-Rabi formula and is shown plotted in Fig. 1 for the special case of $I=3$ and positive magnetic moment. It is most convenient to observe the transitions marked by the short arrow in Fig. 1 and it is obvious that the value of magnetic field at which the Paschen-Back effect of the hyperfine structure begins to be noticeable depends on the magnitude of the nuclear magnetic moment, since the hyperfine structure separation $\Delta\nu$ for two isotopes with the same spin is proportional to the nuclear magnetic moments.

The second of these two methods depends on the ability to convert the atoms of the atomic beam into ions after they have passed through that part of the apparatus required for the radiofrequency magnetic method. For beams of alkali metals, indium, gallium, barium, aluminum, etc., this is easily accomplished by allowing the beam to strike a hot tungsten filament of sufficiently high work function. The ions are then accelerated by an adjustable electric field and allowed to pass through a mass spectrometer magnet. With sufficient resolution the various isotopes can be separated and it is possible to determine whether a particular radiofrequency transition belongs to one isotope or another. Although in principle it is possible, it is certainly not practical to separate two nuclei which differ only because one of them is in an excited state. The isomeric levels of In^{113} and In^{115} are case in point. In these, however, the nuclear spins of the two states will certainly be different.

FIG. 3. Main parts of oven for beams of atomic alkali metals. Alkali azide solution is placed in bottom of cavity in bottom block. Top block is pressed against bottom one with the canal overlapping the cavity.

Once the nuclear spin has been determined as indicated above, it is possible to determine the hyperfine structure $\Delta\nu$, and, incidentally, the nuclear magnetic moment, by observing the Zeeman splitting in successively larger fields until the Paschen-Back effect is strong. The h.f.s. $\Delta\nu$ is then calculated from the observed frequency values and the Breit-Rabi[10] formula. It is frequently possible to make a direct observation of the h.f.s. $\Delta\nu$ by making $\Delta F=\pm 1$ transitions in weak field. Although at the present time it might not seem extremely valuable to the advancement of nuclear physics to know the h.f.s. $\Delta\nu$ of the ground state of a heavy atom like Na^{22} to the precision achievable by the radiospectroscopy method, unless the nuclear magnetic moments are similarly known, a precise value of the h.f.s. $\Delta\nu$ is nonetheless necessary in the determination of the sign of the nuclear magnetic moment by the method employed in the present paper. This method for determination of the sign of the moment depends on being able to measure the very small contribution to the magnitude of the Zeeman splitting that arises directly from the interaction of the nuclear magnetic moment with the applied field. Since this effect is of the order of one thousand times smaller than the Zeeman splitting and, since to such precision the Paschen-Back effect sets in at very small values of magnetic field, it is important not to confuse these two effects of the nuclear moment.

As will be seen later in the discussion of the apparatus, low frequency transitions in the present experiment are restricted to those for which m_F goes from $-F$ to $-(F-1)$. Although the Breit-Rabi formula expresses this transition in closed form, it is perhaps instructive to express it as an expansion about $H=0$ in order to identify the terms referred to above.

Neglecting terms in powers of H higher than two and other small terms, and letting $g_J J\mu_0/h$ equal to 1.400 Mc/sec./gauss, we can write

$$\nu=1.400H/F+1.400\mu_I H/\mu_J F+(1.400)^2(H^2/F^2)(2I/\Delta\nu). \quad (1)$$

The first term is the weak field Zeeman splitting and so predominates over the second term, which is the nuclear magnetic interaction with the applied field, that it can be used directly for the spin evaluation

---

[10] G. Breit and I. I. Rabi, Phys. Rev. 38, 2080 (1931).

without knowing the second term. The third term indicates the beginning of the hyperfine Paschen-Back effect. In practice, the complete Breit-Rabi formula is used, but one has to evaluate the nuclear magnetic moment and the h.f.s. $\Delta\nu$ by a series of successive approximations, obtaining first a rough value of the h.f.s. $\Delta\nu$ from which a sufficiently good value of the nuclear magnetic moment is obtained by comparison with a known stable isotope. This moment value is first assumed positive, and calculations are made with several observed frequency values to find a consistent value of the h.f.s. $\Delta\nu$. Finally, a check can be made of the h.f.s. $\Delta\nu$ directly, since by then the range of high frequency over which one is required to search is sufficiently narrowed to make the process possible. This requirement becomes more evident if it is pointed out that the range of h.f.s. $\Delta\nu$ values for the alkali metals varies from a few hundred megacycles for Li^6 to over ten thousand megacycles for Cs^{137}.

APPARATUS

Figure 2 is an artist's cut-away sketch of the apparatus. Much of the apparatus is similar to that described by Zacharias,[2] in that it was designed for use with rare isotopes in great dilution. The deflecting and refocusing magnets have relatively small ratio of gradient to field in order that a large beam height might be used. Since the beam spreads in height as it leaves the source, the ratio of gradient to field in the refocusing magnet is about half that in the deflecting magnet. Increasing its length to make up for this, offsets to only a small extent the gain from the increased beam height. The slit system is correspondingly wide by comparison with early molecular beam experiments. The widths of the source, the collimator slit, and the detector are all about 0.010 in. The obstacle wire is only a few mils wider than is necessary to shield the hot tungsten filament detector from the undeflected molecules in the beam. Those features which are novel to molecular beam technique are: (1) the oven which is designed to conserve material, (2) the mass spectrometer system which is used to separate the feeble beam of radioactive material from any other ions evaporating from the detector filament, and (3) the Allen type electron multiplier which provides an increase in sensitivity over the best conventional FP54 electrometer by a factor of about 100. The electron multiplier and associated circuits are described in a companion paper by Dr. Hin Lew.

As shown in Fig. 3 the source assembly consists of two simply constructed blocks which are held together by means of a holder containing all heaters, thermocouples, and positioning adjustments. The two lapped surfaces of the blocks when placed in contact must fit well enough so that the main leak out of the oven is through the exit canal which faces the beam detector. The narrow exit canal shown in Fig. 3 is used to diminish the number of atoms diffusing from the slit for a given intensity toward the detector. If we define G as the ratio of atoms diffusing through a hole in an ideal thin wall to the number effusing through the canal for the same forward intensity we have following Smoluchowski[11] and Knudsen,[12] $G=3L/4A$ which is dependent only on the ratio of the length L to the width A. Measurements as shown in Fig. 4 have shown this to be roughly correct provided, of course, that the pressure in the oven is less than 3×10^{-3} mm, in order that the mean free path be larger than the length of the canal.

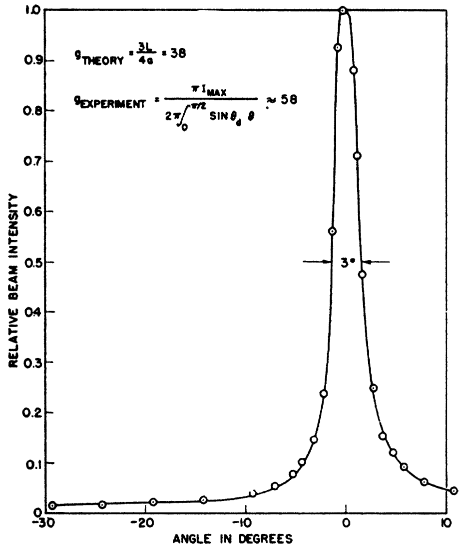

FIG. 4. Relative beam intensity as a function of angle of beam with respect to axis of the canal of the source. For this case, the dimensions of the canal were $0.12\times0.12\times6.3$ mm.

Trials with different kinds of materials for the oven blocks show that many materials are unsuited for minute quantities of alkali metals. One unsuccessful run using stainless steel blocks in which the free sodium was prepared by distillation from a mixture of NaCl and calcium chips displayed two sources of difficulty. The

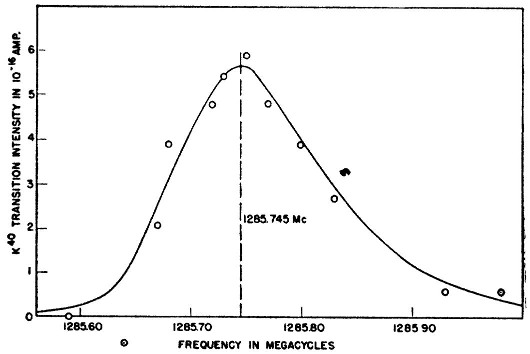

FIG. 5. Resonance curve for the unresolved transitions for the two lines in the K^{40} spectrum given by $F=9/2$, $m_F=\mp1/2\leftrightarrow F=7/2$, $m_F=\pm1/2$ for a steady magnetic field of 5.65 gauss.

---

[11] M. V. Smoluchowski, Ann. d. Physik **33**, 1559 (1910).

[12] M. Knudsen, Ann. d. Physik **31**, 633 (1910).

TABLE I. Observed low frequency transitions for Na^{22}. Corresponding values for the observed frequencies for Na^{23} are included so that by comparison the spin of Na^{22} can be determined.

Na^{23} freq. Mc/sec.	Na^{22} freq.	Ratio	Theoretical ratios
1.10	0.62	0.564	$I=3$ 0.572
1.40	0.80	0.572	$I=4$ 0.444
1.36	0.80	0.588	$I=2$ 0.800
10.37	5.97	0.576	
22.524	13.193	0.586	

calcium chips introduced a large quantity of Na^{23} contamination so that the beam was deficient in Na^{22} for an oven pressure that fulfilled the Knudsen condition. At the end of the run, the Na^{22}, which is an ideal tracer substance for itself, showed that even after washing the oven carefully, a large quantity remained on or in the oven walls, which could be removed only by etching away a considerable thickness of steel.

Trials with other materials such as quartz, nickel, and boron carbide showed either physical absorption or chemical reaction with the oven walls. Of the materials tried, only monel metal seemed satisfactory, and even in this case a small surface area was presented to the metal vapor to minimize such effects.

The surface ionization wire used to ionize the atoms of the beam consists of a flat tungsten ribbon, 0.007 in. wide, mounted flush with the surface of a plane metal guard electrode. These elements are held at an adjustable accelerating voltage with respect to an appropriately slotted plate 0.5 cm away which is held at ground potential. This combination provides excellent ion optics for the mass spectrometer since the field configuration is simple and the initial velocity of the ions is only that due to evaporation from the filament.

The mass spectrometer magnet as indicated in Fig. 2 is a conventional 60° wedge produced by ingot iron pole pieces. It provides an ion path radius of 8 cm in the curved portion. It is to be expected that a mass spectrograph of this sort should have an intensity efficiency of nearly unity, since there are no causes for loss of ions and because the hot tungsten filament converts all incident alkali atoms into ions.

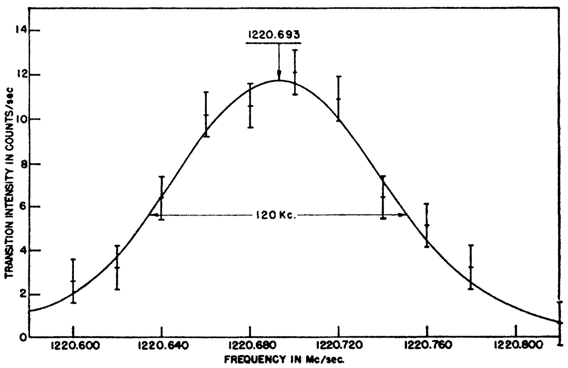

FIG. 6. Transition intensity *vs.* frequency of oscillating field for the two Na^{22} lines $F=7/2$, $m_f=\pm 1/2 \leftrightarrow F=5/2$, $m_f=\mp 1/2$ at a field of 6.17 gauss. One count/sec. is equivalent to about 7 atoms/sec.

The Research Laboratory of Electronics provides a number of signal generators for supplying the radio-frequency current in the homogeneous field "flopping" region, permitting continuous frequency coverage from 0.05 to 2500 Mc/sec. Similarly, there is a variety of frequency meters which includes a laboratory standard, frequently compared with the Bureau of Standards *WWV* 5-Mc/sec. signal.

PROCEDURE AND RESULTS

K^{40}

The first isotope to be tried by the present method was K^{40}, which because of its availability and extremely long radioactive half-life did not require the use of either the long canal oven nor the electron multiplier. A gram of potassium was loaded into a conventional molecular beam oven and heated to give a total beam at the hot wire of 10^9 atoms per second. Of this, about 10^5 atoms per second are of K^{40} and the transition intensities should be a few percent of this. An FP54 electrometer with a sensitivity of 400 ions/sec./mm makes it possible to obtain curves like that of Fig. 5. Without the mass spectrometer, these curves would be superposed on a background which requires some discussion. First, with a common element like potassium, any tungsten filament has bad contamination, which, to be sure, can be largely eliminated, by flashing momentarily to a high temperature and catching the exudate on a liquid air trap surrounding the hot wire. Second, although an obstacle wire provides an almost perfect shadow for protecting the hot wire from direct radiation from the oven, when the deflecting and refocusing fields are energized, there is a background of the order of several tenths of one percent of the beam, the origin of which is discussed more fully in the paper by Dr. Lew. Thus, even though the spin of K^{40} is even and the spins of K^{39} and K^{41} are odd, this large background would make observation of the K^{40} transitions very difficult without the introduction of the mass analysis. It is indeed the product of two discrimination factors, mass analysis and h.f.s. analysis, which permits the experiment to be performed with such small relative abundances.

The $\Delta F=0$ transitions at mass number settings 39 and 40 of the mass spectrometer were observed for various values of the homogeneous magnetic field and, in each case, the only resonance which could be found at the mass 40 position corresponded to $I=4$ for K^{40}, and the transitions for $I=3/2$, which could be clearly identified as being due to K^{39} or K^{41} which the mass spectrometer allowed to fall into the K^{40} position.

With the homogeneous field so low that it was impossible to resolve the various $\Delta F=\pm 1$ lines, search revealed transitions centered at 1285.8 Mc/sec. Sufficient field was then applied to resolve these lines and the observations made which are shown in Fig. 5. These transitions yield a value of $\Delta\nu = 1285.73 \pm 0.050$ which confirms the previous measurement of Zacharias.[2]

Na^{22}

A sample of radiosodium was prepared in the M.I.T. cyclotron by Dr. Eric Clarke in connection with other work, using the $Mg^{24}(d,\alpha)Na^{22}$ reaction. This Na^{22} was then separated from the magnesium by Professor J. W. Irvine, Jr., who exercised the greatest precautions to avoid Na^{23} contamination during the chemical transformations. These procedures can be found in a paper by Irvine and Clarke,[13] who show that the abundance of Na^{22} in the sodium of the sample is about one part in 10^4, and who believe that most of the contamination was contained in the original magnesium target. For the purposes of the present experiment, this abundance provides a reasonable compromise between what is needed for the chemical reactions which must take place in the beam oven and the desire to reduce background as much as possible. Some of the sodium discussed above was converted to NaN_3 by Professor Irvine. A total of 3×10^{-9} mole of $Na^{22}N_3$ along with its carrier was placed in a monel metal oven and the temperature raised to 300°C so that the azide decomposed to give free nitrogen and free sodium. The decomposition of the azide is immediately obvious because of the emission of gas into the chamber surrounding the oven. After the nitrogen is pumped away, the temperature of the oven is lowered to maintain the vapor pressure of the sodium at a proper value.

With the mass spectrometer set for mass number 23, and the homogeneous field set at a value of a few gauss, the $\Delta F = 0$ transition for Na^{23} was observed and the spectrometer reset for mass number 22. A search was conducted at frequencies which correspond to I of 0, 1, 2, 3, 4, 5, 6, and 7. At no frequency other than that for $I = 3$ was any resonance observed. The efficiency of the electron multiplier is such that this peak corresponds to about 40 counts per second. This is just the intensity that was to be expected for the number of states for an atom with $I = 3$, with the known abundance of radiosodium in the sample, and the intensity of the Na^{23} transitions mentioned above. The homogeneous field was then reset to several different values and in each case the transitions at mass 22 were observed at a frequency corresponding to $I = 3$. At each value of the field it was observed that no such transition occurred at mass number 21.7 nor at mass number 22.3. Since the atomic beam apparatus, the detection system, etc., preclude the possibility of this series of transitions being due to any other type of atom or molecule, it is inescapably concluded that the spin of Na^{22} is 3. The appropriate data are shown in Table I.

Observations of this sort were continued up to a field of 170 gauss as shown in Table II. This table also includes values of the h.f.s. $\Delta\nu$ calculated from the data on the assumption, first, that the sign of the magnetic moment is positive and, second, that the sign is negative. From these data either assumption gives reasonably self-consistent results but the results are nonetheless quite different. The magnetic field was then lowered to such a value that the $\Delta F = \pm 1$ lines would be unresolved and a search was made for $\Delta F = \pm 1$ transitions. Only in the neighborhood of 1220.7 Mc/sec. was any resonance observable. In order to obtain an accurate value of the h.f.s. $\Delta\nu$, the field was raised to separate the lines and the data shown in Fig. 6 were taken. The observed line at 1220.694 Mc/sec., when corrected for the small effect due to magnetic field, gives a value of the h.f.s. $\Delta\nu$ of 1220.64 Mc/sec. Thus the sign of the magnetic moment is unambiguously positive. Consideration of the possible sources of error in the high frequency measurements has led us to assign an error of 0.040 Mc/sec. By comparison with the h.f.s. $\Delta\nu$ and magnetic moment μ_I [14] of Na^{23}, we obtain a value of the nuclear magnetic moment for Na^{22} of $+1.746 \pm 0.003$ nuclear magnetons.

It is of interest to consider the amount of radioactive material used in this experiment. For Na^{22} with a 3-year half-life, the 3×10^{-9} mole loaded into the oven was determined by observing the strength of the radioactivity to be 350 microcuries. The total running time

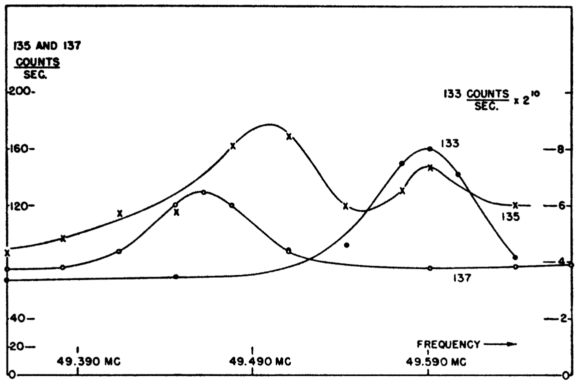

FIG. 7. Resonance curves taken for three different values of mass spectrograph setting. Open circles are for mass 137, dots for 133, and crosses for 135.

TABLE II. Observed low frequency transitions for Na^{22}. Corresponding values for Na^{23} are included. The values of h.f.s. $\Delta\nu$ given are calculated for the two possible signs of the nuclear moment of Na^{22}.

Av. freq. Na^{22}	Av. freq. Na^{23}	h.f.s. $\Delta\nu$ of Na^{22} μ_I pos.	h.f.s. $\Delta\nu$ of Na^{22} μ_I neg.
27.719±0.020	46.038±0.020	1219.2±7.6	1234.2±7.6
30.256±0.010	50.024±0.010	1218.9±3.2	1232.6±3.2
43.126±0.020	69.694±0.020	1216.0±3.4	1225.2±3.4
95.095±0.100	141.900±0.100	1221.9±5.0	1225.4±5.0
	Weighted average of	1218.7±2.2	1229.0±2.2

---

[13] J. W. Irvine, Jr., and E. T. Clarke, J. Chem. Phys. 16, 686 (1948).

[14] H. Taub and P. Kusch, Phys. Rev. 75, 1481 (1949).

TABLE III. Transition frequencies observed with Cs^{133} and Cs^{137} and calculated h.f.s. $\Delta\nu$'s.

Magnetic field in gauss	Av. freq. Cs^{137}	Av. freq. Cs^{133}	h.f.s. $\Delta\nu$ of Cs^{137} μ_I pos.	h.f.s. $\Delta\nu$ of Cs^{137} μ_I neg.
125	44.934 ±0.020	45.092 ±0.020	10,198 ±250	11,301 ±250
136	49.162 ±0.015	49.338 ±0.010	10,130 ±130	11,093 ±130
217	79.842 ±0.026	80.289 ±0.013	10,107 ±80	10,673 ±80
257	95.531 ±0.030	96.198 ±0.030	10,160.9 ±69	10,627.4 ±69
448	175.025 ±0.030	177.105 ±0.030	10,105.9 ±21	10,341.3 ±21
693	289.280 ±0.070	294.873 ±0.090	10,115.1 ±21	10,247.3 ±21
815	352.300 ±0.04	360.560 ±0.04	10,123.5 ±12	10,229.5 ±12
1096	512.125 ±0.050	529.275 ±0.160	10,141.0 ±11	10,207.8 ±11

Weighted average $\Delta\nu = 10,126.5 \pm 7$ Mc/sec.
$\mu_I = 2.837 \pm 0.010$ nuclear magnetons

of the experiment was 16 hours and it was found to have required about 40 microcuries or 4×10^{-10} mole of Na^{22} evaporated from the oven, by observing that the remainder was still in the oven. It is felt that if the running time were reduced by making use of the rapid response of the electron multiplier and photographic recording of data, that it should be possible to perform such an experiment with not much more than 10^{13} atoms. It should thus be possible to work with samples with half-lives as short as are chemically feasible. For instance, 10^{13} atoms with a mean life of 10^3 sec. would amount to $\frac{1}{3}$ curie, which even if lost would not contaminate an apparatus after a few days.

Cs^{137}

A one-millicurie unit of $Cs^{137}Cl$ was obtained from the Isotopes Division of the Atomic Energy Commission. It was found to contain approximately one part Cs^{135} to one part Cs^{137}, as both are fission products and descend from unstable Xe isotopes. A total of 2×10^{-5} mole of $Cs^{133}Cl$ was mixed with 2×10^{-8} mole of $Cs^{137}Cl$ and about 3×10^{-5} mole of NaN_3 and placed in the oven. When in the vacuum the temperature was raised so that the azide decomposed to give free sodium. Because of the higher vapor pressure of Cs, it was displaced from the chloride by the sodium, yielding free cesium.

The $\Delta F=0$ transition of Cs^{133} was then observed, the mass spectrometer tuned for mass number 137, and a search conducted at each frequency corresponding to $I=1/2$ to $I=2\ 3/2$. At no frequency other than that corresponding to $I=7/2$ was any transition observed, but for $I=7/2$ about 60 counts per second were observed. This is just the intensity to be expected from the known abundance ratio and the Cs^{133} intensity observed. It was shown that this transition was not associated with any other odd mass number definitely assigning it to 137. Since the atomic beam apparatus excludes the possibility of observing anything but an alkali atom, the spin of $I=7/2$ may be definitely associated with Cs^{137}.

TABLE IV. Transition frequencies observed with Cs^{133} and Cs^{135} and calculated h.f.s. $\Delta\nu$'s.

Av. freq. Cs^{133}	Av. freq. Cs^{135}	h.f.s. $\Delta\nu$ of Cs^{135} μ_I pos.	h.f.s. $\Delta\nu$ of Cs^{135} μ_I neg.
45.092±0.020	44.996±0.020	9777±240	10,800±240
49.338±0.010	49.235±0.010	9717±100	10,600±100
80.289±0.013	79.998±0.026	9769±80	10,290±80
360.560±0.040	355.155±0.040	9732.1±9	9825±9
408.360±0.040	402.190±0.040	9717.0±7	9795±7

Weighted average $\Delta\nu = 9724.0 \pm 7$ Mc/sec.
$\mu_I = 2.724 \pm 0.010$ nuclear magnetons

The $\Delta F=0$ transition of both Cs^{133} and Cs^{137} were then observed for various magnetic fields up to about 1100 gauss. These data are shown in Table III, also shown are the calculated values of $\Delta\nu$ assuming μ_I first positive and then negative. Consistent results are obtained only for a positive nuclear magnetic dipole moment. As no oscillators were readily available for the frequencies associated with $\Delta F=\pm1$ transitions, the value of $\Delta\nu=10,126.5\pm7$ Mc/sec. is obtained from the $\Delta F=0$ data. By comparison with the $\Delta\nu$ and moment μ_I *** of Cs^{133} we get a nuclear magnetic dipole moment $\mu_I=+2.837\pm0.010$ nuclear magnetons.

Cs^{135}

In order to observe Cs^{135} in the sample of radioisotope obtained from the Isotopes Branch of the Atomic Energy Commission, our experimental procedure was varied slightly from that used for Cs^{137}, since there was considerable residual Cs^{133} at the 135 position of the mass spectrometer. The mass spectrometer was improved somewhat by narrowing the ionizing hot tungsten wire and the receiving slit of the electron multiplier, until the abundance resolution of the spectrometer was better than 1/100 for two mass numbers at 133. The concentration of radio isotope was increased over that for 137 to a concentration of 1/100 so that the Cs^{135} peaks might be clear of background. Figure 7 shows resonance curves taken for three different settings of the mass spectrometer. Without any question there are present three isotopes of the masses 133, 135, and 137 all with spins of 7/2, but with slightly different hyperfine structure and corresponding nuclear magnetic moments. The data of Table IV represent the experimental results and show that, in order to make them self-consistent, it is necessary to assume that the nuclear moment of Cs^{135} is likewise positive. The value of the h.f.s. $\Delta\nu$ can be taken to be 9724.0±8 Mc/sec. and the magnetic moment calculated by comparison

TABLE V. Summary of results.

	Spin	h.f.s. $\Delta\nu$ Mc/sec.	Nuclear magnetic moment μ_I in nuclear magnetons
Na^{22}	3	1220.64±0.04	1.746±0.003
K^{40}	4	1285.73±0.05	−1.290±0.005
Cs^{133}	7/2	9192.76±0.10	2.575±0.007
Cs^{135}	7/2	9724.00±8	2.724±0.010
Cs^{137}	7/2	10,126.50±7	2.837±0.010

*** The value of the nuclear moment of Cs^{133} given by Taub and Kusch (see reference 14) has been recomputed using $I_{133}=7/2$.

with the $\Delta\nu$ and the nuclear moment of Cs^{133} yields 2.724 ± 0.010 nuclear magnetons.

The result, including values for Cs^{133} given by Kusch and Taub,[15] and Bitter[16] are summarized in Table V.

DISCUSSION AND RESULTS

K^{40}

The results previously reported by Zacharias[2] have all been confirmed. In addition, it is of interest to note that the mass assignment to mass 40 is, due to the mass spectrometer, completely unambiguous. In the previous observations this assignment depended on the principle of exclusion applied to all other known alkali metals, and this type of reasoning is never quite as satisfactory as a positive identification.

The value of the h.f.s. $\Delta\nu$ determined in the present experiment completely confirms the negative sign for the magnetic moment of K^{40}. Various writers, chiefly Inglis,[17] have pointed out that with the large spin of 4, one has to invoke nuclear states with outlandishly large values of orbital angular momentum in order to concoct a negative moment. In a private communication, Dr. E. Feenberg has pointed out that, if one assumes *JJ* coupling for the coupling between the proton angular momentum and the neutron angular momentum, it is possible to devise a reasonable level scheme that will give a satisfactorily large value of the negative moment. This may add weight to the type of level scheme recently proposed by Mayer and Teller to account for the so-called magic numbers.

Na^{22}

Na^{22} is the heaviest of the nuclei with equal odd numbers of neutrons and protons for which the spin has been determined. Until the present observation it was thought that these nuclei all had a spin of 1 like the deuteron. Now it is known that B^{10} and Na^{22} depart from this rule. On the other hand, the magnetic moments of B^{10} and of Na^{22} would indicate that these nuclei can be described by the sum of an orbital angular momentum of 2 with an associated nuclear magnetic moment of 1 nuclear magneton and a spin angular momentum of 1 with a magnetic moment of that of the deuteron. We thus have to compare a theoretical value of 1.85 for both with the experimental values of 1.80 for B^{10} and 1.76 for Na^{22}.

The beta-ray spectrum and energy level scheme has been investigated by many observers, the most reliable of which seem to us to be that of Good, Peaslee, and Deutsch[6] who say that the positron emission to an excited state of Na^{22} is allowed but unfavored and is followed by a prompt gamma-ray to the ground state of Na^{22}. Direct transition by positron emission to the ground state of Na^{22} they say has not been observed. In a private communication, Professor Deutsch has said that this latter transition is less than 1 part in 5000 of the observed transition. An angular momentum change of three is sufficient to account for this result. Furthermore, if Gamow-Teller selection rules hold, then it can be concluded that the angular momentum of the excited state of Na^{22} is 2, 3, or 4. Since the positron emission and the gamma-ray are in prompt coincidence it should be possible to observe the spin of this intermediate state of Na^{22} by angular correlation experiments. Professor Deutsch and his co-workers are endeavoring to perform this experiment, which may indeed throw some light on the question of the selection rules in beta-ray theory.

Cs

The most striking feature of the present set of experimental results is a new confirmation of the observation that the addition of two neutrons to a nucleus has very little effect. In the case of these Cs isotopes this effect occurs twice with no change in spin and only a gradual change in the nuclear magnetic moment. It is not expected, however, that the ratios of the h.f.s separations will not be equal to the ratios of the nuclear magnetic moments as found by Bitter[18] in the case of rubidium, since the spins of the cesium isotopes are the same, the magnetic moments are roughly equal so that whatever nuclear volume effects give rise to the small discrepancy in rubidium would not likely be present here. We feel justified in computing the nuclear magnetic moments by the ratio method.

There has recently been considerable discussion of the spin assignment to be made to Cs^{137}, which at present is best summarized by Mitchell and Peacock[8] and by Shull and Feenberg.[19] Since the spin of Cs^{137} is unambiguously 7/2, it is now possible to conclude that the angular momentum of the 153-sec. metastable level of Ba^{137} cannot be as large as 13/2 and still maintain a probability of transition to the ground state which is small compared to the probability of transition to the metastable state. Also it is difficult to see how the spectroscopic determination of the spin of the ground state of Ba^{137} by Benson and Sawyer[20] could be interpreted to give a value of 1/2 instead of 3/2. There remains only the possibility that the theoretical interpretation of the lifetime of the metastable state and of the internal conversion ratio measurements are not adequate to differentiate between angular momentum changes of 4 and 5.

---

[15] P. Kusch and H. Taub, Phys. Rev. **75**, 1477 (1949).

[16] F. Bitter, Phys. Rev. **75**, 1326 (1949).

[17] D. R. Inglis, Phys. Rev. **60**, 837 (1941).

[18] F. Bitter, Phys. Rev. **76**, 150 (1949).

[19] F. B. Shull and E. Feenberg, Phys. Rev. **75**, 1768 (1949).

[20] A. N. Benson and R. A. Sawyer, Phys. Rev. **52**, 1127 (1937).