of U^{238} can take place for energies of the impinging neutrons larger than

$$E' = 5.7 + 2.6 = 8.3 \text{ Mev},$$

where the second term represents the energy carried out by the evaporating neutron from the compound system U^{239} which was calculated as two times the nuclear temperature. The difference between our experimental data and the theoretically expected value of E' is certainly due, in great part, to experimental errors.

As it was pointed out by Bohr, we must expect a similar behavior of the fission cross section in case of Th and Pa. For Th, Bohr gives the following values of the partial cross section for direct fission of Th^{233} and successive fission of Th^{232}

$$\sigma_f = \frac{1}{25}\sigma_0, \quad \sigma_f' = \frac{8}{25}\sigma_0.$$

In the reasonable assumption that the successive fission process of Th^{232} begins to be possible for about the same energy E' of the impinging neutrons as found in case of uranium, we must expect that the mean experimental cross section for fission of Th obtained with the $D+$Li neutrons be

$$1 + 8\alpha' = 1 + \gamma \times 0.13 = 2$$

times larger than the same quantity measured with neutrons of the $D+$Be reactions. Combining the data of Table IV and the result of Section 5, we obtain

$$(1.40 \pm 0.05) \times (1.35 \pm 0.08) = 1.9 \pm 0.2,$$

which seems to agree very well with the theoretical expectation.

Finally we point out that, although our data in case of Pa do not permit any definite conclusion, they seem to indicate that by increasing the energy of the impinging neutrons the fission cross section increases less than the fission cross section of uranium, as we must expect according to the data given by Bohr.

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The Nuclear Magnetic Moments of C^{13}, Ba^{135} and Ba^{137} *

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The nuclear gyromagnetic ratios for C^{13}, Ba^{135} and Ba^{137} have been measured by the molecular beam magnetic resonance method. The signs of the nuclear magnetic moments have also been determined. For C^{13} the g value is 1.402 ± 0.004 and the moment is positive. When this fact is considered with the results of theoretical treatments of the C^{13} nucleus, $I = \frac{1}{2}$ seems to be the preferable value for C^{13}. The resultant nuclear magnetic moment is $+0.701 \pm 0.002$ n. m. The moments for both Ba^{135} and Ba^{137} are positive. The g values are 0.5575 ± 0.0017 and 0.6236 ± 0.0019 respectively. Using $I = \frac{3}{2}$ for both nuclei, the corresponding nuclear magnetic moments are $+0.8363 \pm 0.0026$ and $+0.9354 \pm 0.0029$, respectively. The ratio of the moments is 1.1174 ± 0.0010. All g values are referred to that for Li^7 of 2.1688. Careful examination of the barium beam intensity as a function of homogeneous field strength with constant oscillating field frequency revealed only two resonance minima.

INTRODUCTION

DIRECT experimental data on the nuclear spin and magnetic moment of C^{13} have more than intrinsic interest, for this isotope is one of the light nuclei to which theoretical calculations can be applied with some hope of success. Rose and Bethe[1] have used the Hartree model with Thomas spin-orbit coupling to predict a value of $\frac{1}{2}$ for the nuclear spin while Sachs,[2] on the other hand, obtains the value $\frac{3}{2}$

* Publication assisted by the Ernest Kempton Adams Fund for Physical Research of Columbia University.

[1] M. E. Rose and H. A. Bethe, Phys. Rev. **51**, 205 (1937).
[2] R. G. Sachs, Phys. Rev. **55**, 825 (1939).

by the use of the alpha-particle model and the same type of coupling. Inglis[3] has pointed out that for C^{13} the Larmor coupling term may be as important as, if not more so than, the Thomas term and that if it is taken into account, one obtains $I=\frac{1}{2}$ from either the Hartree or the alpha-particle model. A definite experimental value for the nuclear spin would provide sufficient ground for choice between the theoretical bases of the three calculations, but the only value for this observable is that of Townes and Smythe[4] which was based on measurements of the alternating intensities of lines in an incompletely resolved band spectrum. They selected 3/2 as the best of three possibilities: 1/2, 3/2 and 5/2. Therefore, when Hutchison, Stewart and Urey[5] made available NaCN and KCN in which the abundance of C^{13} was as high as 25 percent, it was considered highly desirable to add to the existing experimental data by applying the molecular beam magnetic resonance method[6-8] to the determination of (a) the nuclear gyromagnetic ratio and (b) the sign of the nuclear magnetic moment of C^{13}. A brief account of the results of these measurements has already appeared;[9] part of this paper will be devoted to an amplification of that report.

Since McLennan and Allen[10] reported hyperfine structure in a number of the arc and spark lines of barium, several workers have used the h.f.s. patterns to evaluate the nuclear spins of the odd isotopes of barium. Schüler and Jones[11] deduced the value $\frac{3}{2}$ for both isotopes from the measurements of Ritschl and Sawyer[12] on the resonance lines of Ba II, $6^2S_{\frac{1}{2}}-6^2P_{\frac{1}{2},1\frac{1}{2}}$. Kruger, Gibbs and Williams[13] repeated intensity measurements on these same lines and concluded that $3/2 \leqq I < 7/2$ with $I=5/2$ as the most probable value. Murakawa[14] made intensity measurements on these and other lines and gives $\frac{3}{2}$ as the value for both isotopes. In 1937 Benson and Sawyer[15] obtained new data from a study of the intensity ratios, the separation of the components from the center of gravity and the nature of the patterns for the $6^2S_{\frac{1}{2}}$, $6^2P_{\frac{1}{2}}$, $7^2S_{\frac{1}{2}}$, 5^3D_3, 6^3P_2 and other terms and they conclude that the spin of each isotope is $\frac{3}{2}$.

Data from the paper by Kruger, Gibbs and Williams have been used by Bethe and Bacher[16] in the Fermi-Segrè-Goudsmit formula to compute the nuclear magnetic moments of the odd isotopes. They give $\mu=+1.06$ and $+0.82$ for Ba^{135} and Ba^{137}, respectively. In their tables of nuclear magnetic moments, however, both moments appear as 1 (or 0.9 with $I=\frac{3}{2}$). Part of the inability to resolve the two moments is undoubtedly due to the fact that the even isotopes of barium result in a very heavy structureless component that almost obscures the h.f.s. components due to the odd isotopes, and part is due to the impossibility of resolving the splitting of the $6^2P_{\frac{1}{2},1\frac{1}{2}}$ terms by optical methods. The problem thus posed is one which the molecular beam magnetic resonance method is particularly well able to solve, for it does not respond to effects due to nuclei of zero spin or zero moment and its resolution is well beyond that of spectroscopy. In addition, barium possesses a $^1S_0$ ground state which means that atomic beams can be used instead of molecular beams. Thus the complications which arise from intramolecular interactions and possible rotational states are eliminated and the results can be expected to give some check on the theory of the method.

Experimental

The apparatus and the experimental technique which were used to obtain the results set forth in this paper are those which have been described by others[6,7] and that description need not be repeated here except for a review of the procedure

[3] D. R. Inglis, Phys. Rev. **56**, 1175 (1939).
[4] C. H. Townes and W. R. Smythe, Phys. Rev. **56**, 1210 (1939).
[5] C. A. Hutchison, D. W. Stewart and H. C. Urey, J. Chem. Phys. **8**, 532 (1940).
[6] I. I. Rabi, S. Millman, P. Kusch and J. Zacharias, Phys. Rev. **55**, 526 (1939).
[7] S. Millman, Phys. Rev. **55**, 628 (1939).
[8] A. F. Stevenson, Phys. Rev. **58**, 1062 (1940).
[9] R. H. Hay, Phys. Rev. **58**, 180 (1940).
[10] J. C. McLennan and E. Allen, Phil. Mag. **8**, 515 (1929).
[11] Kallman and Schüler, Ergeb. d. exact. Naturwiss. **11**, 134 (1932).
[12] R. Ritschl and R. A. Sawyer, Zeits. f. Physik **72**, 36 (1931).
[13] P. G. Kruger, R. C. Gibbs and R. C. Williams, Phys. Rev. **41**, 322 (1932).
[14] K. Murakawa, Sci. Pap. Inst. Phys. Chem. Research, Tokyo **18**, 304 (1932).
[15] A. N. Benson and R. A. Sawyer, Phys. Rev. **52**, 1127 (1937).
[16] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 222 (1936).

used to determine the sign of the magnetic moments. Some information will be given, in addition, about the compounds used for the C^{13} measurements, the way in which the C^{13} resonance minimum was identified and the difficulties encountered in detecting the barium beams.

The resonance minima can be obtained in either of two ways: by observing beam intensity as a function of the strength of the homogeneous field with the frequency of the oscillating field held constant (constant frequency method), or by observing the beam intensity as a function of the oscillating field frequency with the homogeneous field strength held constant (constant field method). In each case, the curves must be taken in pairs to avoid errors due to the end effects of the oscillating field. One member of the pair is taken with the applied fields in what has been designated the Normal direction, while the other is taken with the fields in the opposite, or Reverse, direction. Millman[7] has pointed out that this procedure for obtaining accurate g values also gives information about the sign of the nuclear magnetic moment to be associated with a minimum, for the direction in which the minima shift with reversal of the applied field directions depends upon the sign of the moment. If it is to low field values, when the fields are changed from Normal to Reverse, the moment is positive. In the case of C^{13}, the curves were all obtained by the use of the constant frequency method since it is usually more convenient from an experimental point of view. In the case of the barium isotopes, however, it was found that when this method was used the minima were so narrow and the end effects so small that the shifts in minima were obscured by the errors caused by the demagnetization procedure. For that reason, and also because the ratio of the g values is much more accurately calculated from curves obtained by the constant field method, some of the barium curves were taken in this way.

C^{13}

The cyanides procured from Professor Urey's laboratory were formed by titrating a condensate of HCN with KOH and NaOH. The chemical purity of all of the substances used was at least equal to that of the KOH, the maker's analysis of which is given below:

Na_2CO_3	2.5	percent
Cl	0.010	"
SO_4	0.005	"
PO_4	0.005	"
SiO_2 and NH_4OH ppt. (SiO_2, Al_2O_3, Fe_2O_3)	0.020	"
N	0.001	"
Fe	0.002	"
Heavy metals	0.003	"

The molecular beams were formed by heating the cyanides in iron ovens to about 700°C and the greatest experimental difficulties arose from the excessive dissociation of the NaCN at that temperature and its tendency to condense on the oven slit jaws. The cure lay in careful drying and moderate preheating under vacuum.

The surface ionization detector used in these experiments has so far been successfully used to detect only the alkali metals, gallium, indium and barium. Fortunately, it will detect the alkali elements when they are combined with other elements to form compounds stable enough to give molecular beams. This circumstance makes possible the application of the molecular beam magnetic resonance method to the measurement of the nuclear g value of elements which are not directly detected by the surface ionization detector but which combine stably with the alkalis. Kusch, Millman and Rabi[17] have reported such measurements on the nuclei of chlorine, aluminum, nitrogen and other elements. As they point out, the correct assignment of observed resonance minima to nuclei is usually possible only if a given minimum is observed with two or more compounds which have nothing in common but the element of immediate interest. In the case of C^{13} only two compounds were available. Had either of these been obtainable in large amounts, it would have been desirable to use that one to form a third compound such as LiCN or RbCN in the hope that it, too, would form usable molecular beams and thus make easier the task of identifying new minima. Since this course was impracticable because of the limitation of supply, the following procedure was substituted: The first step was to study the intensity of the KCN beam as a

[17] P. Kusch, S. Millman and I. I. Rabi, Phys. Rev. **55**, 1176 (1939) and subsequent papers.

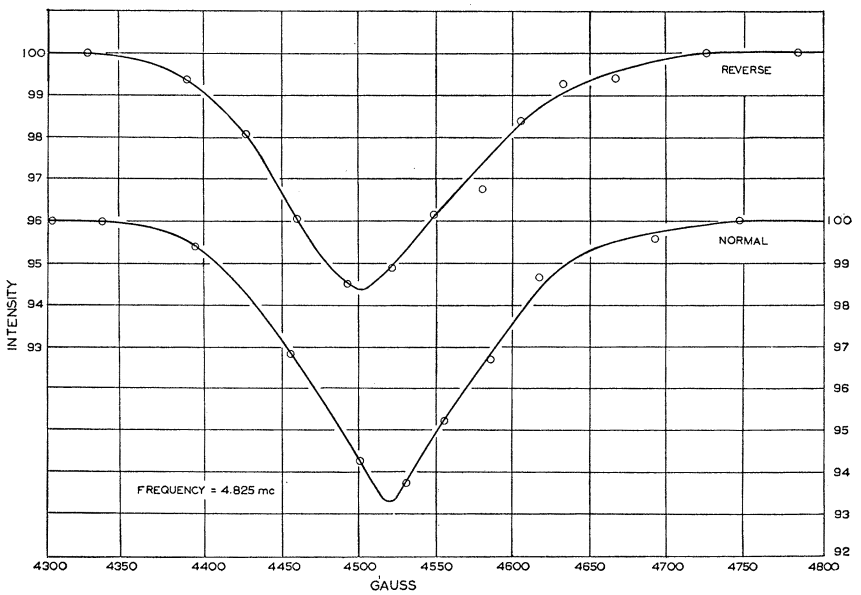

FIG. 1. Beam intensity as a function of homogeneous field strength for beams of KCN enriched in C^{13}. The lower curve is taken with the applied fields in the Normal direction, the upper, with them in the opposite, or Reverse, direction.

function of the homogeneous field strength with the frequency being kept fixed for each curve. Only three resonance minima were seen, two of which were at once identified as being due to K^{39} and N^{14}. The third, curves for which are given in Fig. 1, did not correspond to any g value recorded in this laboratory so it was tentatively assigned to C^{13}. Next, the intensity of the NaCN beam was plotted as a function of the field strength but the new minimum turned out to be so close to that due to Na^{23} that it was visible only as an asymmetry on the high field side of the latter. Two of the curves for the region near the Na^{23} minimum are shown in Fig. 2. They indicate that the asymmetry is not due to end effects of the oscillating magnetic field, for it does not change from the high field side of the minimum to the low with reversal of the field directions. The third step was to obtain beams of KCN and NaCN containing only the normal amount (1.06 percent) of C^{13}. Beam intensity curves of normal KCN showed only two resonance minima, that for K^{39} and that for N^{14}. The new one assigned to C^{13} was absent. The two curves given in Fig. 3 are typical of the normal NaCN beam intensity for the region near the Na^{23} minimum. They reveal no trace of the asymmetry to be seen in the curves of Fig. 2.

The problem is to identify the new minimum with the nucleus of C^{13}. It can be done by eliminating all other alternatives. The chance that a chemical impurity is responsible is negligible, for the NaCN and the KCN used were prepared from substances free from all but the merest traces of impurities important in these experiments. Nor can the new minimum be due to C^{12} for that nucleus possesses no magnetic moment. Even if it did, its minimum should appear in the beams of ordinary KCN in which only two minima were found. There remains the possibility that the minimum is due to the moment arising either from the rotation of the molecules as a whole or from the decoupled rotation of a group, such as CN, common to both molecules. To be valid, the first hypothesis would require that the moments of inertia of NaCN and KCN molecules be equal, which is unlikely. The second would demand complete decoupling of such a rotation from the rotation of the molecule as a whole, which would seem rather difficult to accomplish at the low fields used (not over 5000 gauss). In any case, it is interesting to repeat the computation which

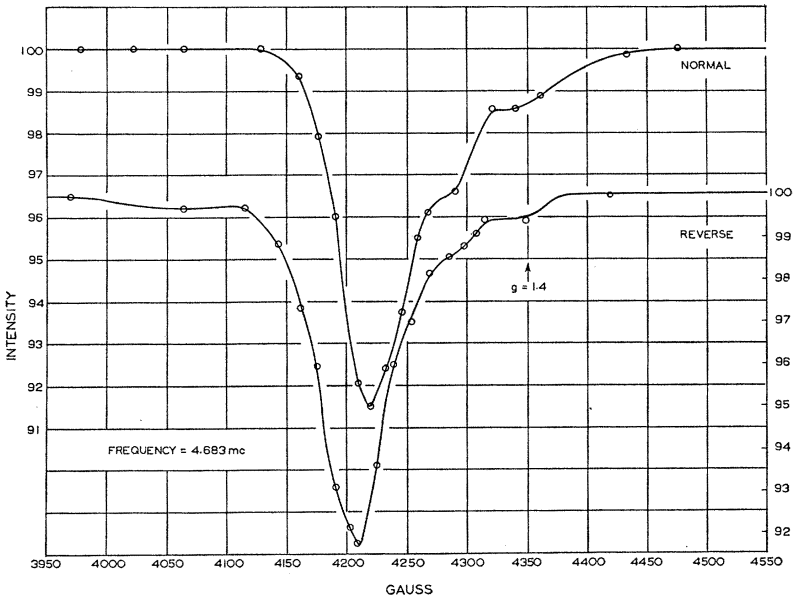

FIG. 2. Beam intensity as a function of homogeneous field strength for beams of NaCN enriched in C^{13}. The main resonance minimum is due to Na^{23}. The asymmetry caused by the C^{13} nucleus is indicated by the arrow marked "$g=1.4$."

Kusch, Millman and Rabi[17] have made of the probable nuclear magnetic moment of the CN group, in which they postulated that the observed minimum is due to such a rotation. The separation of the carbon and the nitrogen atoms is known[18] to be 1.15×10^{-8} cm in the HCN molecule and it seems reasonable to assume this same value for the KCN molecule. The moment of inertia of the CN group then is $I=14.2\times10^{-40}$ g cm^2. From the relation, $Jh/2\pi=(2IkT)^{\frac{1}{2}}$, the most probable value for the rotational angular momentum for an oven temperature of 1000°K comes out to be $19h/2\pi$. The new minimum gives a g value of about 1.4, which leads to a magnetic moment of $19\times1.4=26.6$ nuclear magnetons. By observing the deflection of the molecular beam caused by a single inhomogeneous field one can form an estimate of the approximate magnetic moment of the KCN molecule. Experimental values are about one nuclear magneton, not even the same order of magnitude as the result calculated above. Thus, rotational states may be ruled out as possible causes of the new minimum observed in the enriched KCN beam. At very low values of the homogeneous field, minima yielding g values which are integral multiples of the true g value have been observed in molecular beams for nuclei the spins of which are greater than $\frac{1}{2}$. The new minimum observed in these experiments cannot be attributed to any such phenomenon, however, for it was seen at high field values while both theory and experimental observation show that multiple g value minima are not. This exhausts the alternatives and, therefore, the third minimum seen in the beams of enriched KCN was assigned to the C^{13} nucleus.

[18] H. Sponer, *Molekulspectren* (J. Springer, 1935).

Ba^{135} and Ba^{137}

The barium metal used in these experiments was procured from Eimer and Amend and was over 99 percent pure when bought. The chief impurity was oxygen. As a check, the barium residues left in two ovens after use were analyzed spectroscopically. Silicon, iron, tin, lead and aluminum were found to be present to less than 0.09 percent while strontium, calcium, zinc, sodium and potassium were not found. The beams were formed in the usual manner by heating the metal in an iron oven to about 950°C.

Barium has an ionization potential of 5.16 volts and, therefore, the surface ionization

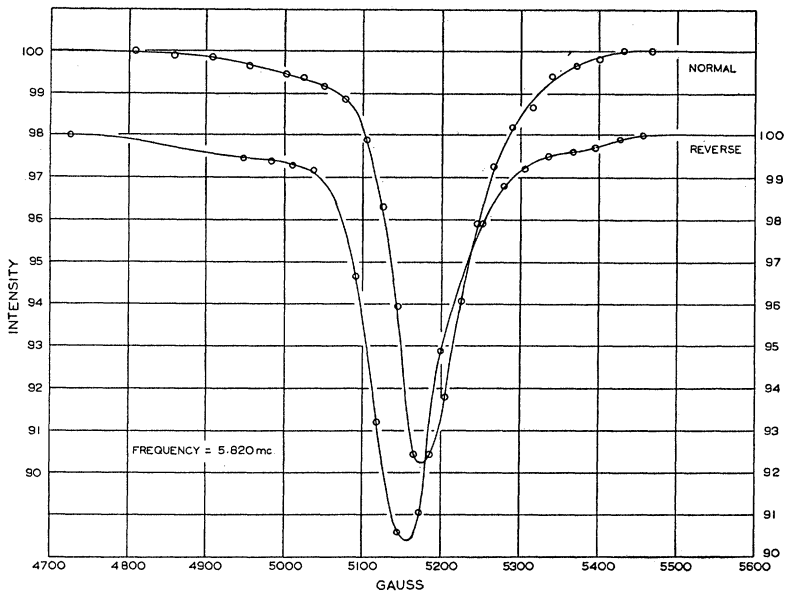

FIG. 3. Beam intensity curves of the region near the Na^{23} minimum for beams of NaCN containing only normal amounts of C^{13}. Note the absence of the asymmetry so noticeable in the curves of Fig. 2.

detector should not detect it.[19,20] An oxide-coated filament will detect elements such as sodium and lithium, whose ionization potentials are higher than 4.5 volts, the work function of tungsten, but it will not work for barium, probably because BaO is an efficient agent for lowering the apparent work function of the filament. However, in 1936 Guthrie[21] reported that, contrary to current theories of surface ionization, barium could be detected by a clean tungsten filament if the latter were kept at sufficiently high temperatures. The precautions which he listed as necessary to secure satisfactory detection were observed with success, although considerable experience was required before the best results were obtained.*

The barium beams were usually very unsteady. Figure 4 gives some idea of the degree of unsteadiness, especially when it is noted that each point on the curve is the average of several observations any one of which may be a rather bad fluctuation. This unsteadiness was due in part to the high filament temperature which caused the barium beam to be superimposed on a background of continuously fluctuating positive

[19] J. B. Taylor, Zeits. f. Physik **57**, 242 (1929).

[20] R. G. J. Fraser, *Molecular Rays* (Cambridge University Press, 1931).

[21] A. N. Guthrie, Phys. Rev. **49**, 868 (1936).

* Guthrie made some calculations based on the geometry of his apparatus, the measured positive ion current and the kinetics of the beam which led him to state that when it was above 2000°K, the filament converted all of the neutral barium atoms striking its surface into positive ions. The temperature, estimated from electrical conditions, at which the filament was maintained for detection in my experiments was at least 2000°K. If the filament, after some steady use as a barium detector, was used for lithium, the detection of which requires an oxide coat, it was found impossible to maintain the required coat and detect lithium until after the filament had been subjected for two or three hours to a temperature well above that used for barium detection and dangerously close to that which experience has shown will burn it out in a short while. This odd behavior of the tungsten was inexplicable until it was recalled that barium in combination with oxygen is a very efficient agent for lowering the apparent work function of tungsten. During the time it had been used as a barium detector, the tungsten had adsorbed a coat of barium with which the oxygen promptly combined to form barium oxide which, in turn, had the opposite effect on the work function of the filament to that necessary for the detection of lithium. The phenomenon was described to Dr. J. A. Becker of the Bell Telephone Laboratories who immediately recalled the results of some unpublished measurements he had made on the ability of tungsten to ionize barium atoms. He found that the conversion from neutral atoms to positive ions at the surface of a hot tungsten wire was by no means equal to 100 percent but was only a third to a half that much. The methods used both in these and in Guthrie's experiments to determine filament temperatures are too inexact to permit one to say that my experience invalidates his statements. It does suggest, however, that tungsten is an inefficient barium detector and requires higher oven pressures and greater beams than would have been necessary with a more efficient detector, and that the phenomena of barium detection will bear more examination.

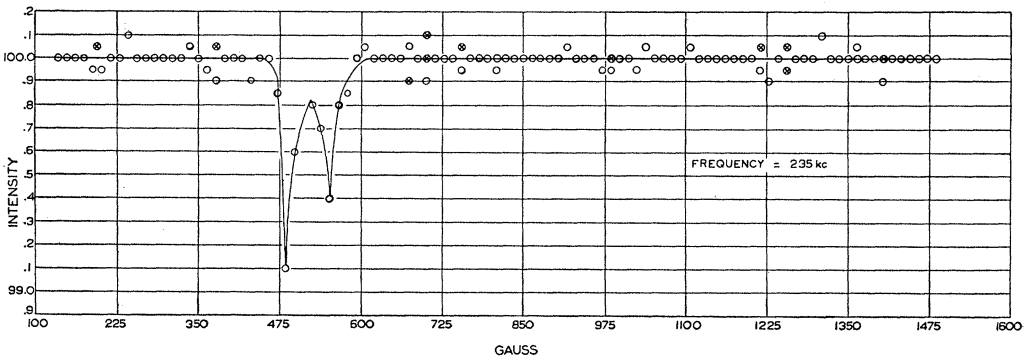

FIG. 4. An exploratory curve giving the barium atomic beam intensity as a function of homogeneous field strength. The frequency of the oscillating field was held constant at 235 kilocycles. Points marked ⊗ are those taken as repeat observations to make sure that beam decreases indicated by several of the original points were not due to resonance minima but only to beam fluctuations.

ion current. Another contributing factor was the excessive oven pressure necessitated by the inefficient detection process. Unprecedented quantities of barium, to judge by experience with other elements and compounds, had to be evaporated from the ovens to maintain a beam of even moderate intensity for a matter of six or seven hours.

To add to these difficulties, it was discovered that the power of a single inhomogeneous field to deflect atoms from the beam was extremely small, being, even with a collimating slit 0.02 mm wide, a matter of less than six percent. This figure set an upper limit not only to the depth of any minimum but also to the sum of the depths if more than one minimum were seen, and meant that extreme care would have to be taken in order that no small minima would be skipped. Each reading of beam intensity represents the difference between the intensity with the oscillating field off and that immediately after with the field on. Furthermore, each of these readings is the average of as many as eight or ten separate observations. In most cases, the averaging was done mentally, but it is interesting to note that what was done with the aid of an adding machine indicated that the extra time and labor involved in the use of the machine was not justified by any significant gain in accuracy.

RESULTS

C^{13}

The resonance curves shown in Fig. 1 are typical of those obtained from observations of the intensity of beams of KCN enriched in C^{13}. They were obtained by the use of the constant frequency method. The shift of the minimum with reversal of the direction of the applied fields is easily seen and indicates that the nuclear magnetic moment of C^{13} is positive. The proper field value to be used in calculating the apparent g value is, of course, the average of the minimal abscissae of the two curves. Data from seven pairs of such curves were used in the formula,

$$g=\frac{1.3122\times10^{-3}\times f}{H}$$

to calculate the apparent g value for C^{13}. H is the value of the homogeneous field at resonance and f is the resonant frequency. Referred to the value 2.1688[22] for Li^7, it is, $g=1.402\pm0.004$ in units of $e/2Mc$.

Ba^{135} and Ba^{137}

Figure 4 is a curve of beam intensity *vs.* homogeneous field strength for the barium atomic beam. The frequency of the oscillating magnetic field was 235 kc and the range of homogeneous field values was such as to cover g values from 0.2 to 6 in units of $e/2Mc$. There are only two resonance minima, nor did more than these two appear in any of the other exploratory curves that were taken. The total range of g values covered was from 0.1 to 9 and for the low values, care was taken to keep the strength of the oscillating magnetic field inversely proportional to the g value in accordance

[22] Private communication from S. Millman and P. Kusch.

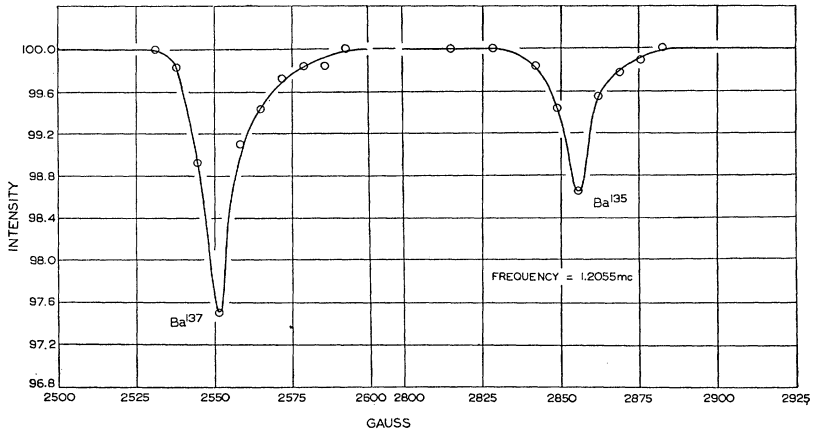

FIG. 5. Beam intensity as a function of homogeneous field strength for the two resonance minima seen in the barium atomic beams. Note their extreme narrowness and the very slight asymmetry.

with the theory of the method. When these two minima were examined in detail, curves such as those of Figs. 5 and 6 were obtained. The curves of Fig. 5 were taken in rapid succession so that they might be used to get the ratio of the minimal ordinates. They also indicate the narrowness of the barium curves, their half-widths being about 0.47 and 0.42 percent (approximately 4.5 kc), respectively. Curves taken in this manner were not satisfactory for determining the signs of the moments, for they showed only slight asymmetries and no shifts of minima with reversal of the directions of the applied fields, the shifts being so small as to be totally obscured by the demagnetization error in the homogeneous field values. Therefore, the curves of Fig. 6 were taken with the frequency of the oscillating field as the independent variable and the strength of the homogeneous field constant. These curves are as narrow as the others (half-widths = 0.43 percent) but not only do the asymmetries due to oscillating field end effects appear more pronounced but also the shifts in minima with reversal of applied field directions are easily discerned. In addition to those of Fig. 6, two more curves were taken for the shallower minimum. The first was with an oscillating field of about 5 gauss and the second with a field four times that strength. The minimum for the second curve was located 1.4 kc higher in frequency than was that for the first, a clear indication that the moment associated with this minimum is positive. The sign of the moment for the deeper minimum is determined by the curves of Fig. 6 to be positive.

The following apparent nuclear g values were calculated from all of the curves taken: 0.5537 $\pm$0.0016 for the smaller minimum and 0.6193 $\pm$0.0018 for the other. The units in both cases are those of the fraction $e/2Mc$, and both values are referred to the value 2.1688 for Li^7. The ratio of the g values was determined from the curves of Fig. 6 and is 1.1174$\pm$0.0010. Four values for the ratio of the decreases in intensity at resonance were obtained: 1.60, 1.84, 2.09 and 1.76, the average being 1.82. Three sets of figures for the abundances of the isotopes of barium are given in Table I, those of Aston,[23] of Sampson and Bleakney[24] and of Nier.[25] The only two isotopes whose abundance ratio is near 1.8 are Ba^{137} and Ba^{135}; their ratio is given in the final column of the table.

The evidence and the arguments upon the basis of which the g value 0.5537 was assigned to Ba^{135} and the value 0.6193 to Ba^{137} are as follows:

(1) The resonance minima observed for the nuclei of Li^6 and Li^7, B^{10} and B^{11}, Cl^{35} and Cl^{37} and Rb^{85} and Rb^{87} show that for each of these isotope pairs, the ratio of the decrease in intensity at resonance is fairly closely proportional

[23] F. W. Aston, Proc. Roy. Soc. **134**, 571 (1932).
[24] M. B. Sampson and W. Bleakney, Phys. Rev. **50**, 456 (1936).
[25] A. O. Nier, Phys. Rev. **54**, 275 (1938).

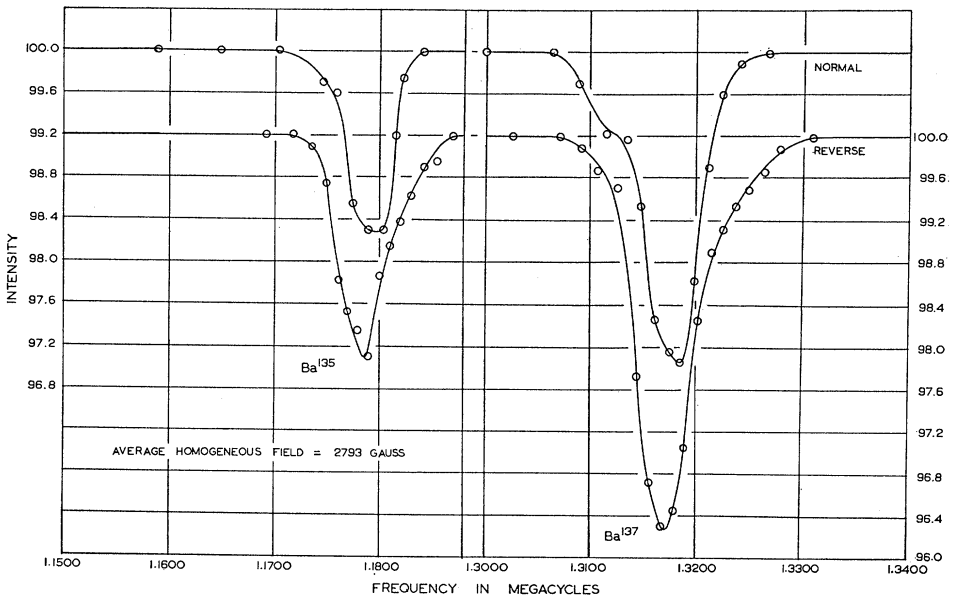

FIG. 6. Beam intensity as a function of oscillating field frequency with the homogeneous field strength held constant. These curves indicate clearly the positive sign of the nuclear magnetic moment to be associated with each minimum.

to the isotope abundance ratio. These minima were observed in the intensity curves of molecular beams and are therefore subject to broadenings and complications arising from intramolecular interactions. What holds for them can confidently be expected to hold for the much narrower minima seen in the barium atomic beam intensity curves, for none of the molecular complications are present.

(2) It is very unlikely that Ba^{138} is responsible for either of the observed minima, for if it were, its abundance is such that a single inhomogeneous field would deflect past the detector more than 6 percent of the total beam. This argument does not apply to the isotopes Ba^{134} and Ba^{136} but there remains a third which is applicable to all the even isotopes.

(3) If either of the two minima is attributed to one of the even isotopes, we can reasonably expect to find a third minimum for the odd isotope with which no minimum would then be associated. Furthermore, in view of the abundance ratios, it should be of about the same depth. No such minimum was found. This bears out the view of spectroscopists that the h.f.s. patterns seen in barium spectra are caused by the two odd isotopes. From these patterns it has been established that these isotopes have equal spins and so nearly equal moments that it is impossible to resolve them optically.

DISCUSSION

Precision

An estimate of the precision of these measurements of nuclear g values involves consideration of errors in four factors: the constant factor, 1.3122×10^{-3}, into which are lumped all atomic physical constants appearing in the formula for calculating g values;[6] the frequency of the oscillating magnetic field; the homogeneous field calibration constant by means of which the potential drop across a shunt in the windings is converted to field strength, and the location of the minimal abscissae for the resonance curves. As far as these results are concerned, errors in the constant factor are entirely negligible. Frequencies were measured with a heterodyne frequency meter capable of an accuracy of 0.005 percent and the oscillator used was stable to better than 500 cycles for the time necessary to take a curve, so that errors in frequency are small enough to be ignored. The homogeneous field was not measured absolutely but was

TABLE I. *Abundance of isotopes.*

Mass Number	130	132	133	134	135	136	137	138	Ratio 137/135
Abundance, Aston	—	—	—	—	5.9	8.9	11.1	74.2	1.88
Abundance, Sampson and Bleakney	0.16	0.015	0.1	1.72	5.7	8.5	10.8	73.1	1.90
Abundance, Nier	0.101	0.097	—	2.42	6.59	7.81	11.32	71.66	1.71

calibrated so that the g values were referred to that for Li^7. Before and after the measurements on C^{13} and again after the measurements on the barium isotopes, the resonance frequency of the Li^7 minimum was determined for field values which covered the working range of the homogeneous field. Since the field strength is indicated in practice by the potential drop across a shunt in the field windings, this calibration procedure results in a curve of a certain factor (by means of which millivolt readings are converted to field strength) as a function of millivolts drop across the shunt. The experimental points were scattered due to small variations in demagnetization procedure, but the greatest deviation from the average for any one field value was 0.3 percent and the average deviation for all points and all three calibrations was 0.15 percent. Location of the minimal abscissae introduces more error than does any other factor since this error depends on such things as the nature of the curves, their widths and the number of points close to the minimum. A conservative estimate of the precision of the location of the C^{13} minimal abscissae is 0.3 percent. The curves for the barium isotopes are considerably sharper than those for C^{13} but, except for those of Fig. 6, no greater precision of location is claimed for them because the minima are, in too many cases, dependent on one point. If all of the errors are combined in the usual way, the value for the probable error is 0.3 percent.

It will be noticed that the precision of the ratio of the g values of the barium isotopes is somewhat greater than that of the values themselves. This ratio was determined from the curves of Fig. 6 and such a determination eliminates errors due to all but very short time changes in the homogeneous field and reduces practically to the measurement of two frequencies.

A correction must be applied to the observed nuclear g values to take into account the diamagnetic property of the atom concerned. Lamb's expression[17] is,

$$\bar{H}/H=0.320\times10^{-4}\times Z^{4/3},$$

where Z is the atomic number and $\bar{H}/H$ is the correction to the external magnetic field H due to the field $\bar{H}$ created at the nucleus by the rotating electrons. For C^{13} this correction is less than 0.04 percent and so is neglected, but for the barium isotopes it amounts to 0.7 percent and the corrected g values become: $g=0.5575\pm0.0017$ for Ba^{135} and $g=0.6236\pm0.0019$ for Ba^{137}, both in units of $e/2Mc$.

The nuclear magnetic moment of C^{13}

When the nuclear spin is known, the measured g value leads at once to the value of the nuclear magnetic moment. Unfortunately, the spin of C^{13} is not known with certainty. Townes and Smythe, who made the only experimental determination, give the value $\frac{3}{2}$ as more probable than the value $\frac{1}{2}$, but their data do not rule out the latter altogether. Of the three theoretical predictions that have been made, it is possible to eliminate Sachs' at once, for, by his theory, the negative moment of the extra neutron is added algebraically to the smaller moment due to the rotational angular momentum and the resultant nuclear magnetic moment is always negative, in direct contradiction to the experimentally determined fact that it is positive. Inglis[26] has pointed out additional support for the view that the nuclear spin of C^{13} is $\frac{1}{2}$. It is derived from the empirical rule that all hitherto known magnetic moments of odd nuclei lie between the limits for the simple single-particle model. The corresponding limits for the gyromagnetic ratio, g, would be, for the odd neutron case,

$$\mu_p/I<g<-\mu_p/(I+1),$$

where $\mu_p=-1.96$, is the lower limit of the

[26] D. R. Inglis, Phys. Rev. **58**, 577 (1940).

neutron moment. The observed g value for C^{13} exceeds the upper limit of 1.30 only slightly for $I=\frac{1}{2}$ but it exceeds it by a factor of two or more for any higher spin value. These arguments strongly support the acceptance of $\frac{1}{2}$ as the most likely value for the nuclear spin of C^{13}, but it must be emphasized that they do not by any means constitute an experimental determination which is contradictory to that of Townes and Smythe.

If the nuclear spin is taken as $\frac{1}{2}$, the value for the nuclear magnetic moment of C^{13} is, $\mu=+0.701\pm0.002$ nuclear magneton, a value in fair agreement with that calculated by Rose and Bethe which is $\mu=+1.05$ n.m.

In view of the recent experimental work by Staub, Tatel and Stevens[27,28] on the anomalous scattering of neutrons by helium, and Dancoff's[29] theory of spin-orbit coupling in He^5 which involves tensor forces of a magnitude sufficient to account for the anomaly, it is unnecessary to make a choice between the calculations of Rose and Bethe and those of Inglis, for both are likely incomplete and not to be relied on too heavily. Not that Rose and Bethe's value for the magnetic moment will be affected much by the addition of the tensor forces, however, for they will determine which level of the ^2P state lies lower while the g value formula will give a moment which is largely due to neutron spin.

The nuclear magnetic moments of Ba^{135} and Ba^{137}

The spectroscopic determinations of the spins of the odd isotopes of barium have already been reviewed. The work of Benson and Sawyer was undertaken to resolve the uncertainty created by the value 5/2 which Kruger, Gibbs and Williams gave. Since the former authors' value 3/2 is based on evidence additional to that presented by the latter authors and is supported by two other independent determinations, it will be used in this paper. Accordingly, the nuclear magnetic moments of the two odd barium isotopes are: $\mu=+0.8363\pm0.0026$ nuclear magneton for Ba^{135} and $\mu=+0.9354\pm0.0029$ nuclear magneton for Ba^{137}. Their ratio is 1.1174 ± 0.0010.

The Fermi-Segrè-Goudsmit formulas have been used to calculate these nuclear magnetic moments from the observed separation factors for the $6^2S_{\frac{1}{2}}$ and $6^2P_{\frac{1}{2}}$ terms. Bethe and Bacher's results, calculated with $I=5/2$, have already been given; with $I=3/2$ they can be corrected to $\mu=+0.954$ and $+0.74$ n.m. for Ba^{135} and Ba^{137}, respectively. That the alteration in moment with change in spin is slight is due to the fact that μ is proportional to $I/(I+\frac{1}{2})$. The g value, on the other hand, is much more sensitive since it is proportional to $1/(I+\frac{1}{2})$. Since they can be compared to experimental values, g values calculated from barium h.f.s. separations for spin values of 5/2 and 3/2 should yield additional information to support the choice of $I=3/2$. Table II gives the results of such a calculation:

TABLE II. *g values.*

	$6^2S_{\frac{1}{2}}$	$6^2P_{\frac{1}{2}}$
$\Delta\nu$ (obs)	0.260 cm^{-1}	0.038 cm^{-1}
$(I+\frac{1}{2})g$	1.25	0.95
g (for $I=3/2$)	0.62	0.42
g (for $I=5/2$)	0.42	0.32
g (obs) Ba^{135}	0.56	0.56
g (obs) Ba^{137}	0.62	0.62

The spin value $I=3/2$ gives a g value in satisfactory agreement with the experimental values, but $I=5/2$ results in a g value too discordant to be acceptable, especially when the $6^2S_{\frac{1}{2}}$ term alone is considered. The values obtained from the $6^2P_{\frac{1}{2}}$ state are low but, according to Goudsmit,[30] this is to be expected. The g values calculated from spectroscopic data emphasize the lack of resolution of the optical method, for one cannot distinguish a separate value for each isotope. The extension in resolution offered by the molecular beam magnetic resonance method is strikingly demonstrated by the ease with which the two minima observed in the barium atomic beams could be resolved. The potentialities of the method are shown by the following figures. Assume that the half-widths of the minima will not exceed 5 kc and that it is desirable to have the minima separated by a frequency increment

[27] H. Staub and W. E. Stevens, Phys. Rev. **55**, 131 (1939).
[28] H. Staub and H. Tatel, Phys. Rev. **58**, 820 (1940).
[29] S. M. Dancoff, Phys. Rev. **58**, 326 (1940).
[30] S. Goudsmit, Phys. Rev. **43**, 636 (1933).

equal to about two or three times the half-widths. The convenient limit of the homogeneous field strength is about 6500 gauss; at that field the Ba^{137} minimum would occur at about 3 mc and it would be quite easy to observe another minimum 15 kc away, provided both were as sharp as are the two barium minima. This separation corresponds to a g value ratio of 1.0050. In other words, had the moments differed by only 0.5 percent, the difference would have been detected with certainty and without extraordinary demands upon experimental technique.

One of the most important parts of the work on the barium isotopes is the search that was made for resonance minima other than the two that were seen. It has already been pointed out that barium is of particular interest. In the first place, it is possible to work with atomic beams and what minima there are should be ideally sharp uncomplicated by anything but the Maxwellian velocity distribution of the beam itself. In the second place, most of the isotopes are of even mass number, and, within the limits mentioned below, the experiments should provide some test of the assumption that the even isotopes are of zero spin or do not possess a magnetic moment. Rough calculations indicated that the smallest moment which would allow either of the inhomogeneous fields alone to deflect atoms past the detector would be of the order of 0.3 nuclear magneton, which means that with a spin of $\frac{3}{2}$, the smallest g value likely to be seen would be of the order of 0.2. To be on the safe side, the lower limit to which the search was extended was $g=0.1$ in units of $e/2Mc$. The upper limit was 9. The theory of the method requires that the strength of the oscillating field be inversely proportional to the g value. Consequently, for one of the exploratory curves, the oscillating magnetic field strength was increased so as to be always at least the proper magnitude, the starting point being the field strength necessary to give a reasonably sharp minimum for Ba^{137}. The positive ion current from the detecting filament was never steady but was always subject to random fluctuations whose magnitudes were such as to make it unwise to claim that minima of depth less than 0.5 percent of the beam intensity could have been recognized. A caution altogether natural in view of the detection difficulties which were encountered dictates that the smallest recognizable minimum be put at about half that due to Ba^{135}. These experiments establish, then, that if any of the isotopes of barium other than 135 and 137 possesses a gyromagnetic ratio, that ratio must be less than 0.2 and is likely less than 0.1, in units of $e/2Mc$, or else the g value is due to an isotope of less than 2.5 percent abundance. Further evidence that only the isotopes 135 and 137 have gyromagnetic ratios is to be found in the close correspondence which exists between the sum of the decreases in beam intensity at resonance and the decrease in intensity caused by a single inhomogeneous field. The latter decrease sets an upper limit to the sum of the decreases due to resonance minima. It was difficult to measure with any accuracy because of the lateral shift of the beam which arose from the diamagnetism of the barium beam, but it amounted to about 6 percent. The sum of the two decreases at resonance was observed in one curve to be 5.8 percent leaving 0.2 percent to be accounted for by another isotope, by inaccuracies in the determination of the single field decrease or by the fact that the strength of the oscillating field was greater than necessary and thus caused a diminution of the intensity decrease at resonance. Of these three probable explanations the second and third are by far the more important.

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