The Molecular Beam Resonance Method for Measuring Nuclear Magnetic Moments

The Magnetic Moments of $_3Li^6$, $_3Li^7$ and $_9F^{19}$*

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(Received January 20, 1939)

A new method of measuring nuclear or other magnetic moment is described. The method, which consists essentially in the measurement of a Larmor frequency in known magnetic fields, is of very general application and capable of the highest precision in absolute and relative measurements. The apparatus consists of two magnets in succession which produce inhomogeneous magnetic fields of oppositely directed gradients. A molecular beam of the substance to be studied possesses a sigmoid path in these magnets and is focused on a suitable detector. A third magnet which produces a homogeneous field is placed in the region between the two deflecting magnets. In this strong homogeneous field the nuclear moments are decoupled from other nuclear moments and from rotational moments of a molecule in a $^1\Sigma$ state, and precess with their Larmor frequency $\nu=\mu H/hI$. An oscillating field perpendicular to the homogeneous field produces transitions to other states of space quantization when the frequency of this field is close to ν. If such transitions take place the molecule is no longer focused on to the detector by the subsequent inhomogeneous field and the observed intensity diminishes. The application of the method to the molecules LiCl, LiF, NaF and Li_2 is described. The nuclear moments of Li^7, Li^6 and F^{19} were found to be 3.250, 0.820 and 2.622 nuclear magnetons, respectively.

THE magnetic moment of the atomic nucleus is one of the few of its important properties which concern both phases of the nuclear problem, the nature of the nuclear forces and the appropriate nuclear model. According to current theories the anomalous moment of the proton is directly connected with the processes from which nuclear forces arise. The question whether the intrinsic moments of the proton and neutron are maintained within the nucleus is part of the problem of two and multiparticle forces between nuclear constituents. With regard to the atomic model it is clear that the nuclear angular momentum does not alone suffice to fix the nature of the wave functions which specify the state of the nucleus. The magnetic moment, on the other hand, is sensitive to the relative contributions of spin and orbital moment and, with the advance of mathematical technique, suffices to decide between the different proposed configurations.

In the light of these considerations it is particularly desirable that nuclear moments be known to high precision because small effects may be of great importance. A case in point is that of $_3Li^7$; according to the calculations of Rose and Bethe[1] the contribution of the orbital motions to the moment of this nucleus is about 10 percent of the total moment. The rest is contributed by the intrinsic proton moment. If the nuclear moment were known to only 10 percent, the importance of this datum would be greatly diminished.

In two letters to this journal,[2, 3] we reported briefly on a new precision method of measuring nuclear moment, and on some results. In this paper we shall give a more detailed account of the method, apparatus and results.

METHOD

The principle on which the method is based applies not only to nuclear magnetic moments but rather to any system which possesses angular momentum and a magnetic moment. We consider a system with angular momentum, J, in units of $h/2\pi$, and magnetic moment μ. In an external magnetic field H_0 the angular mo-

* 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] I. I. Rabi, J. R. Zacharias, S. Millman and P. Kusch, Phys. Rev. **53**, 318 (1938).
[3] I. I. Rabi, S. Millman, P. Kusch and J. R. Zacharias, Phys. Rev. **53**, 495 (1938).

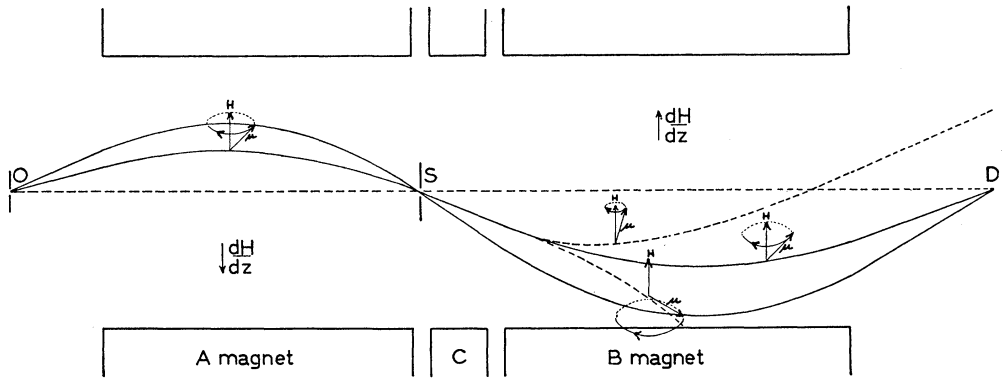

FIG. 1. Paths of molecules. The two solid curves indicate the paths of two molecules having different moments and velocities and whose moments are not changed during passage through the apparatus. This is indicated by the small gyroscopes drawn on one of these paths, in which the projection of the magnetic moment along the field remains fixed. The two dotted curves in the region of the B magnet indicate the paths of two molecules the projection of whose nuclear magnetic moments along the field has been changed in the region of the C magnet. This is indicated by means of the two gyroscopes drawn on the dotted curves, for one of which the projection of the magnetic moment along the field has been increased, and for the other of which the projection has been decreased.

mentum will precess with the Larmor frequency, ν, (in revolutions per sec.) given by,

$$\nu = \mu H_0 / Jh. \tag{1}$$

Our method consists in the measurement of ν in a known field H_0. The measurement of ν is the essential step in this method, since H_0 may be measured by conventional procedures. Using Eq. (1) we obtain the gyromagnetic ratio. If, in addition, the angular momentum, J, of the system is known, we can evaluate the magnetic moment μ. In its present state of development our method is not suitable for the measurement of J.

The process by which the precession frequency ν is measured has a rather close analog in classical mechanics. To the system described in the previous paragraph, we apply an additional magnetic field H_1, which is much smaller than H_0 and perpendicular to it in direction. If we consider the initial condition such that H_1 is perpendicular to both the angular momentum and H_0, the additional precession caused by H_1 will be such as to increase or decrease the angle between the angular momentum, J, and H_0, depending on the relative directions. If H_1 rotates with the frequency ν this effect is cumulative and the change in angle between H_0 and J can be made large. It is apparent that if the frequency of revolution, f, of H_1 about H_0 is markedly different from ν, the net effect will be small. Furthermore, if the sense of rotation of H_1 is opposite to that of the precession, the effect will also be small. The smaller the ratio H_1/H_0 the sharper this effect will be in its dependence on the exact agreement between the frequency of precession, ν, and the frequency f.

Any method which enables one to detect this change in orientation of the angular momentum with respect to H_0 can therefore utilize this process to measure the precession frequency and therefore the magnetic moment. The general method here outlined includes not only the magnitude but also the sign of the magnetic moment since the direction of precession depends on whether the magnetic moment vector is parallel or antiparallel to J.

The precise form of the initial conditions previously described is not important and we may consider H_1 initially at any angle ϕ with the plane determined by H_0 and J but still perpendicular to H_0. In fact, according to quantum mechanics, we must consider the initial conditions of an ensemble of systems with a definite projection of J on H_0 as uniformly distributed over ϕ. This only means that some systems will increase and other systems will decrease their projections in the direction of H_0.

In practice it is frequently more convenient to use an oscillating field H_1 rather than a rotating field. Although the situation is not quite as clear as for the rotating field, it is reasonable to

expect that the effects will be similar if the oscillating field is sufficiently small. A simple calculation shows that no change in the magnitude of the projection of J on H_0 will occur unless the frequency of oscillation is close to the frequency of precession. The sign of the moment does not affect any processes when a pure oscillating field is used, since it may be considered as the superposition of two oppositely rotating fields. Hence no information as to the sign of the moment can be obtained when this type of field is substituted for a rotating field.

Although the reorientations of the system under the combined influence of H_0 and H_1 may be detected in a number of ways,[4] the most delicate and precise is that of molecular beams.

The arrangement used in our experiment is shown schematically in Fig. 1. A stream of molecules coming from the source, O, in a high vacuum apparatus is defined by a collimating slit, S, and detected by some suitable device at D. The magnets, A and B, produce inhomogeneous magnetic fields, the gradients of which, $d|H|/dz$, are indicated by arrows. When these magnets are turned on, molecules having magnetic moment will be deflected in the direction of the gradient if the projection of the moment, μ_z, along the field is positive, and in the opposite direction if μ_z is negative. A molecule starting from O along the direction OS will be deflected in the z direction by the inhomogeneous A field and will not pass through the collimating slit unless its projected moment is very small or it is moving with very high speed. In general, for a molecule having any moment, μ_z, and any energy, $\frac{1}{2}mv^2$, it is possible to find an initial direction for the velocity of the molecule at the source such that the molecule will pass through the collimating slit. This is indicated by the solid lines in the diagram. If d_A denotes the deflection at the detector from the line OSD suffered by the molecule due to the A field alone, it may be expressed by:

$$d_A=(\mu_z/2mv^2)(d|H|/dz)_A G_A.$$

The deflection in the B field will be in a direction opposite to that in the A field and is given by:

$$d_B=(\mu_z/2mv^2)(d|H|/dz)_B G_B.$$

The factors, $(d|H|/dz)_A G_A$ and $(d|H|/dz)_B G_B$, depend only on the geometry of the apparatus and can be adjusted to have the same value. Thus if a molecule of any velocity has the same μ_z in both deflecting fields it will be brought back to the detector by the B field. A simple consideration shows that when the fields A and B are properly adjusted the number of molecules which reaches the detector is the same whether the magnets A and B are on or off. The molecular velocity distribution is also the same.

Magnet C produces the homogeneous field H_0. In addition, there is a device, not pictured in Fig. 1, which produces an oscillating field perpendicular to H_0. If the reorientation which we have described takes place in this region the conditions for deflecting the molecules back to D by means of the B magnet no longer obtain. The molecule will follow one dotted line or the other depending on whether μ_z has become more positive or has changed sign. In fact, if any change in orientation occurs, the molecule will miss the detector and cause a diminution in its reading. We thus have a means of knowing when the reorientation effect occurs.

Since most of the systems in which one is interested have small angular momenta ($<10h/2\pi$) the classical considerations given above have to be reconsidered from the point of view of quantum mechanics. The reorientation process is more accurately described as one in which the system, originally in some state with magnetic quantum number, m, makes a transition to another magnetic level, m'. An exact solution for the transition probability for the case where H_1 rotates and is arbitrary in magnitude was given by Rabi.[5] For the particular case of $J=\frac{1}{2}$ we have,

$$P_{(\frac{1}{2},-\frac{1}{2})}=\frac{\sin^2\theta}{1+q^2-2q\cos\theta}\sin^2\pi tf(1+q^2-2q\cos\theta)^{\frac{1}{2}},$$

where $P_{(\frac{1}{2},-\frac{1}{2})}$ is the probability that the system, originally in the state $m=\frac{1}{2}$ is found in the state $m=-\frac{1}{2}$ after a time t, q the ratio of the frequency of revolution f to the frequency of precession $\nu[\nu=\mu(H_0^2+H_1^2)^{\frac{1}{2}}/Jh]$, and $\tan\theta=H_1/H_0$.

For an oscillating field, in the limit where $H_1/H_0\ll 1$, and in the neighborhood of $f=\nu$ this

[4] C. J. Gorter, Physica 9, 995 (1936).

[5] I. I. Rabi, Phys. Rev. 51, 652 (1937).

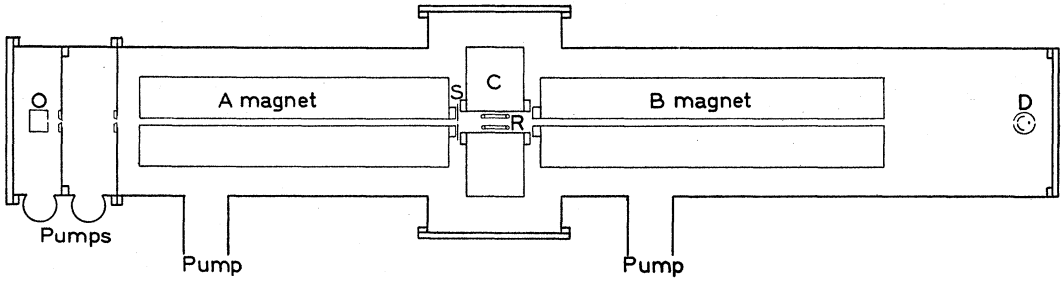

FIG. 2. Schematic diagram of apparatus.

formula becomes:

$$P_{(\frac{1}{2},-\frac{1}{2})}=\frac{\Delta^2}{(1-q)^2+\Delta^2}\sin^2\pi tf[(1-q)^2+\Delta^2]^{\frac{1}{2}}, \quad (2)$$

where $\Delta=H_1/2H_0$, one-half the ratio of the amplitude of the oscillating field to the static field, and the other symbols retain their meaning. For spins higher than $\frac{1}{2}$ the general formula given by Majorana[6] applies, and

$$P_{(\alpha,\ m,\ m')}=(\cos\tfrac{1}{2}\alpha)^{4J}(J+m)!$$
$$\times(J+m')!(J-m)!(J-m')!$$
$$\times\left[\sum_{\nu=0}^{2J}\frac{(-1)^\nu(\tan\frac{1}{2}\alpha)^{2\nu-m+m'}}{\nu!(\nu-m+m')(J+m-\nu)!(J-m'-\nu)!}\right]^2, \quad (3)$$

where α is defined through $P_{(\frac{1}{2},\ -\frac{1}{2})}=\sin^2(\alpha/2)$. That is, we calculate α for a system which has the same μ/i but with a spin of $\frac{1}{2}$ and subject to the same field, and use it in Eq. (3).

The orders of magnitude involved can be seen from a simple example: consider a system with spin $\frac{1}{2}$ and a moment of 1 nuclear magneton in a field of 1000 gauss and an oscillating field of 10 gauss amplitude. We assume that the system is moving at a speed of 10^5 cm per second which is of the order of thermal velocities, and set $t=l/v=10^{-5}l$. The resonance frequency is

$$\frac{uH}{hi}=\frac{(0.5\times10^{-23})(10^3)}{(6.55\times10^{-27})(\frac{1}{2})}\sim1.5\times10^6 \text{ cycles per sec.},$$

which fortunately is in a very convenient range of radiofrequencies. To make the $\sin^2$ terms a maximum at $q=1$ we set

$$\pi\times10^{-5}l\times1.5\times10^6\times0.5\times10^{-2}=\pi/2.$$

Solving for l, we obtain $l=6.6$ cm, which is a very convenient length for the oscillating field.

The theoretically simplest systems to which these ideas may be applied in the study of nuclear moment are atoms which are normally in a state with electronic angular momentum equal to zero. If the electronic J is not zero, the interaction of the nuclear spin with the electronic angular momentum is of the order of magnitude of its interaction with the applied field H_0. Moreover, the electronic magnetic moment is so much larger than nuclear moment that the deflections in the A and B fields will be almost entirely due to this electronic moment and the apparatus will accordingly be insensitive to changes in nuclear orientation. Resort must therefore be had to atoms in a state $J=0$ or to molecules in a $^1\Sigma$ state in which all electronic angular momentum is neutralized to the first order. These considerations do not preclude the study, with these methods, of atoms with electronic angular momentum, as such, but rather point out that they are not the most suitable systems for the investigation of nuclear magnetic moment.

As elementary calculations show, the interactions between the nuclear moments of the nuclei in a molecule in a $^1\Sigma$ state and the other angular momentum vectors, such as molecular rotation, are of the order of magnitude of 100 gauss or less. External fields of a few thousand gauss will therefore decouple all the nuclear spins from each other and from the molecular rotation to such a degree that they may be regarded as free. The other interactions will result in a fine structure of constant or decreasing width as H_0 is increased. These effects on the precision can, therefore, be reduced to any assigned value merely by working at suitably high field.

[6] E. Majorana, Nuovo Cimento 9, 43 (1932).

Apparatus

The apparatus (Fig. 2) is contained in a long brass-walled tube divided into three distinct chambers, each with its own high vacuum pumping system. The source chamber contains the oven which is mounted on tungsten pegs. By means of a screw the mount may be moved, under vacuum, in a direction perpendicular to the beam axis. Stopcock grease and Apiezon *Q* on the screw preserve vacuum even when the screw is turned. The interchamber contains no essential parts of the apparatus, but provides adequate vacuum isolation of the receiving chamber from the gassing of the heated oven, by means of a narrow slit on each end of the chamber. These slits may be moved under vacuum in a manner similar to the oven mount. The receiving chamber contains most of the essential parts of the apparatus: the two deflecting magnets, *A* and *B*, the magnet, *C*, which produces the constant field, the radio-frequency oscillating field, *R*, the collimating slit, *S*, and the 1-mil tungsten filament detector, *D*.

The *A* and *B* fields are electromagnets of the type described by Millman, Rabi and Zacharias[7] and are 52 cm and 58 cm long, respectively. The gap is bounded by two cylindrical surfaces, one convex of radius 1.25 mm, and the other concave of radius 1.47 mm. The gap width in the plane of symmetry, defined by the axes of the two cylindrical surfaces, is 1.0 mm. The nature of the field obtained is approximately the same as that produced by two parallel wires with centers 2.5 mm apart and carrying current in opposite directions. Each magnet has four turns of copper windings; current is supplied by a 3000-ampere-hour, 2-volt storage cell. A current of 300 amperes in the windings yields a field of over 12,000 gauss and a gradient of about 100,000 gauss/cm in the gap.

The *C* magnet, which produces the homogeneous field, is made of annealed Armco iron and is of conventional design. It is wound with 12 turns of $\frac{3}{16}''$ square copper rod to which $\frac{3}{16}''$ copper tubing has been soldered for cooling purposes. Insulation between turns, and between the windings and the magnet, is provided by mica. The pole faces, separated by a gap of $\frac{1}{4}''$, are 10 cm long and 4 cm high. A field of about 23 gauss is realized in the gap per ampere of current in the exciting coils.

In mounting the magnets in the apparatus, care must be taken to avoid regions of weak, rapidly changing fields between magnets. Such regions cause transitions between quantum states of the various magnetic moments associated with the molecule and prevent good refocusing of the beam by the *B* field. Although the gradient in the *B* field is necessarily in a direction opposite to that in the *A* field, the magnetic fields in the planes of symmetry of the two magnets are in the same direction and parallel to that in the *C* magnet. The magnets are placed as close to each other as the windings will permit. Moreover these windings are completely hidden from the "view" of the molecular beam by mounting slabs of iron as extensions on both ends of the *C* magnet and on the ends of the *A* and *B* magnets facing the *C* magnet. This arrangement insures a fairly strong field along the entire path of the molecular beam where changes in the over-all magnetic moment of the molecule affects its position at *D*, i.e., from the beginning of the *A* field to the end of the *B* field, and thus limits transitions between the quantum states to the region of the *R* field, where they may be controlled and studied.

The oscillating field, *R*, consists of two $\frac{1}{8}''$ copper tubes, 4 cm long, carrying current in opposite directions. These tubes are flattened to permit their insertion between the pole faces of the *C* magnet when a space of about 1 mm between the tubes is left for the passage of the beam. The plane defined by the centers of these tubes is horizontal and is adjusted to be closely the same as the planes of symmetry of the *A* and *B* magnets. These tubes are supported by heavy copper tubing through which electrical and water connections may be made outside the apparatus.

The magnetic field, H_1, produced by a current in the tubes is about 2 gauss/amp. and is approximately vertical and therefore at right angles to the field H_0 produced by the *C* magnet. The high frequency currents in the tubes are obtained by coupling a loop in series with them

[7] S. Millman, I. I. Rabi and J. R. Zacharias, Phys. Rev. **53**, 384 (1938).

to the tank coil of a conventional Hartley oscillator in which an Eimac 250 TL tube is used. The frequencies used for these experiments range from 0.6 to 8 megacycles. The currents producing the oscillating field may be varied from 0 to 40 amperes; the higher currents are more easily obtained at low frequencies.

Procedure

A preliminary line-up of magnets, slits and detector is made by optical means while the apparatus is assembled. If this line-up is sufficiently good, a beam may be sent through the apparatus and a more precise line-up made by means of a triangulation process utilizing the property of rectilinear motion of the molecules in the beam.

The A and B magnets have knife edges at both ends which overlap the gap on the side of the convex pole face by known amounts and extend above the gap into the region above the magnet by a known amount. Since it is impossible to sight through the gaps with a telescope, the preliminary optical line-up is made by sighting on the extensions of the edges in the region above the magnets. In this way it is possible to adjust the plane of symmetry of magnet A to coincide with that of magnet B. The optical line-up is sufficient for this purpose, since no very great precision is needed for this adjustment. It is also possible to adjust optically the lateral position of the magnets as well as the slits and detector to permit a beam to pass through the magnet gaps. The C magnet is lined up so that the median plane of its gap coincides with the centers of the gaps of the A and B magnets. The two wires which produce the radiofrequency field, R, are suspended from a brass plate which is mounted on top of the vacuum chamber, and are so constructed that the width of the assemblage is only very slightly less than the width of the gap in the C magnet. The field R is then arranged in the gap so that it does not short to the poles of the C magnet. Since the width of the gap between the two wires is greater than that of the available working gap in the A and B magnets, this line-up is sufficient for the field R.

A sample of the molecular compound, the magnetic moments of whose constituent nuclei are to be determined, is placed in an oven. The oven is completely closed except for a slit about 0.03 mm wide. It is heated by means of spiral tungsten heaters passed through the oven block and electrically insulated from it by means of quartz tubing. When the temperature of the oven is sufficiently high so that the sample has a vapor pressure of the order of 1 mm of Hg a beam may be observed at the detector and a more precise line-up may be initiated.

In the present apparatus the B magnet is permanently fixed inside the vacuum chamber and all other line-up operations are made with respect to it. By suitable movements of the oven, the collimating slit, and the detector, the beam is shifted until it is cut by each of the two fiduciary knife edges on the B magnet in turn. From a knowledge of the distances separating the various elements involved in a cut-off, it is possible to set the beam parallel to the plane defined by the two edges and to ascertain the distance of the beam from that plane. The only measurements that must be made during this line-up process are the readings of detector positions by means of a calibrated tele-microscope. By successive movements of the oven, collimating slit and detector the beam can be translated parallel to itself by any desired amount. It is thus brought into a position at which one would like to have the plane of the edges of the A field. This field is then moved, in a manner similar to that described for the motion of the oven mount, until its fiduciary edges cut the beam. This operation sets the plane defined by the edges on the A field parallel to the corresponding plane of the B field and at a predetermined distance from it. The beam is then translated to a position approximately midway between these planes.

The experimental criterion which determines the exact position of the beam is that the weakening of a molecular beam at the detector by the A and B fields taken separately must be equal. This may easily be accomplished by a lateral displacement of the beam, since for any such displacement the gradient increases in one of the fields and decreases in the other. When this criterion is satisfied the intensity of the refocused beam with a current of about 300 amp. in the windings of each of the two inhomogeneous

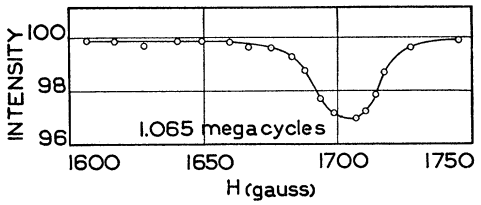

FIG. 3. Resonance curve of the Li^6 nucleus observed in LiCl.

fields is about 95 percent of the beam observed in the absence of fields.

As has been pointed out, the refocusing condition obtains only when there is no change in the space quantization of any of the moment vectors associated with the molecule. If weak fields occur in the region between the A and B fields, transitions may occur. The refocusing becomes good only when the C field itself is fairly large, and the intensity of the refocused beam is an increasing function of the C field up to a value of about 500 gauss. The resonance minima to be described subsequently are usually observed at fields larger than 1000 gauss.

Because the amplifier is not completely shielded from the oscillator and because the steady deflection of the galvanometer associated with the amplifier due to the oscillator is a function of the frequency, observations are made of the beam intensity as a function of the magnetic field, H_0, when the frequency is held fixed. Curves relating the beam intensity to the field H_0, taken for $_3Li^7$, $_3Li^6$ and $_9F^{19}$ are shown in Figs. 3, 4 and 5.

MAGNETIC AND FREQUENCY MEASUREMENTS

Since the value of the magnetic moment of any nucleus is calculated from an observed magnetic field and an observed frequency it is essential that these quantities be known to a high degree of precision. The frequency of the oscillating magnetic field is determined to better than 0.03 percent by measuring the frequency of the oscillator with a General Radio Type 620A heterodyne frequency meter. It was found that the frequency of the oscillator varied by no more than 0.01 percent during the time required to obtain data on one resonance curve ($\sim$15 minutes).

A calibration of the magnetic field of the homogeneous C magnet in terms of the current through the exciting coils was made in the usual way by measuring the ballistic deflection of a galvanometer when a flip coil was pulled from the magnetic field. The galvanometer was calibrated by the use of a 50-millihenry mutual inductance, good to $\frac{1}{2}$ percent. Several flip coils were constructed in this laboratory by winding various types and sizes of insulated wire on carefully measured brass spools. Errors in the magnetic field due to uncertainties in flip coil areas are probably not greater than 0.2 percent.

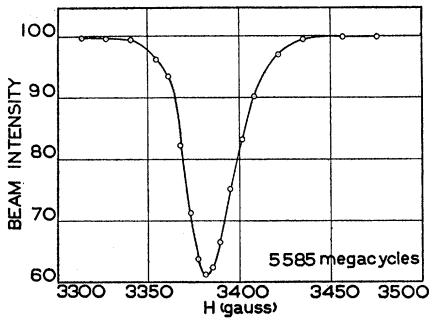

FIG. 4. Resonance curve of the Li^7 nucleus observed in LiCl.

A type K potentiometer was used to measure the potential drop across a shunt in series with the C magnet windings. The same shunt was used both in the calibration and in subsequent work, thereby eliminating the necessity of knowing its resistance accurately.

It is important that the magnetic field always return to the same value for a given magnetizing current. It was found that when a definite, reproducible procedure was used for demagnetizing the homogeneous field and for bringing it up to any state of magnetization, this condition was fulfilled to better than 0.1 percent.

A considerable variation in the value of the mutual inductance was observed, apparently depending on the humidity. The absolute value of the magnetic field is indeterminate to about 0.5 percent due to the uncertainty in the value of the mutual inductance and uncertainty in the areas of the flip coils. This, of course, introduces a corresponding uncertainty in the absolute values of the magnetic moments.

RESULTS

The first nuclei to be studied by this method were $_3Li^6$, $_3Li^7$ and $_9Li^{19}$ in the LiCl, LiF, NaF and

Li_2 molecules. The resonance minima which are obtained are shown in Figs. 3, 4 and 5. For each nucleus the f/H values corresponding to the resonance minima are constant to a very high degree for wide variations of frequency. This shows that we are dealing with a change of nuclear orientation and not with some molecular transition, since such a transition would not possess a frequency proportional to H. A representative sample of the results is shown in Table I. The constancy of f/H also shows that our method of calibration of the C magnet yields accurate results, at least for relative values of the homogeneous field.

The nuclear g is obtained from the observed f/H values by use of the formula

$$g=\frac{4\pi}{e/Mc}\cdot\frac{f}{H}=1.3122\times10^{-3}\frac{f}{H},$$

which follows immediately from Eq. (1) if the magnetic moment μ is measured in units of $eh/4\pi Mc$, the nuclear magneton, and $f=\nu$. The specific charge of the proton in electromagnetic

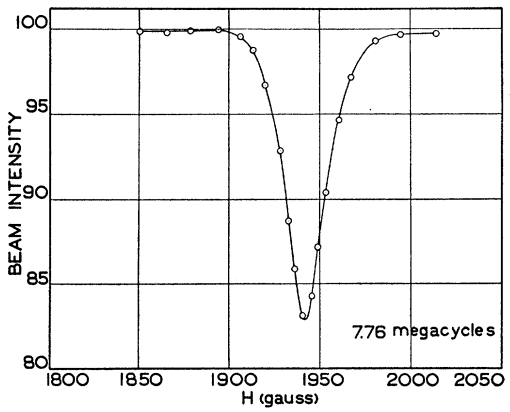

FIG. 5. Resonance curve of the F^{19} nucleus observed in NaF.

units, e/Mc, is obtained directly from the value of the Faraday[8] (9648.9 e.m.u.) and the atomic weight[9] of hydrogen (1.0081). Expressed in this form our experimental results do not depend upon any inaccuracies in e, h or m/M, the ratio of the electronic mass to the mass of the proton. The nuclear spins of Li^6 and Li^7 are known from atomic beam measurements,[10, 11] and that of F^{19} from band spectra.[12] The nuclear moments are obtained directly by multiplying g by i. The nuclear g's, the spins and the magnetic moments are listed in Table II. The values here given are about 0.5 percent lower than, and are to supersede, those published in the preliminary report.[3] The differences are due to the use of a more trustworthy mutual inductance in the calibration of the magnetic field and to an error in the value of the constant $4\pi/(e/Mc)$ previously used. The identification of the resonance minimum with a particular nucleus is made by using the same element in more than one molecule. For example, two of the resonance minima, observed for each of the molecules,[13] LiCl, LiF and Li_2 have f/H values which are the same in all three cases. These must be attributed to the nuclei of the two isotopes of lithium. Since Li^7 is about 12 times as abundant as Li^6 and since the intensity drop at resonance for one of these minima is as much as 60 percent of the refocused beam, this minimum can only be assigned to Li^7. No minimum is definitely assigned to a nucleus unless it has been observed in at least two different molecules.

The accuracy of the nuclear moment values depends solely on a knowledge of the magnetic field, H, at which the Larmor frequency associated with the nuclear magnetic moment is equal to the frequency of the oscillating field. The absolute moment values depend upon the absolute calibration of magnetic standards and cannot at present be taken to be better than 0.5 percent. The relative moment values, on the other hand, do not depend on such standards but merely on the accuracy of the shape of the, magnetization curve for the homogeneous field on the reproducibility of a definite field with the same current in the exciting coils of the homoge-

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[13] The Li_2 molecules are obtained by heating lithium metal to about 1000°K. At vapor pressures of one mm the beam contains about 0.5 percent molecules. The lithium atoms have magnetic moments of the order of one Bohr magneton and suffer such large deflections in the A field that they do not reach the detector. The problem of working with alkali molecules of the type of Li_2 is solely one of obtaining enough intensity for the molecular beam. The atoms do not interfere with the experiment in any way.

neous field, on the location of a minimum in the resonance curve and on the assumption that any form of interaction tending to broaden the resonance curve and not considered in the simple theory will introduce no asymmetry into the curve. The criterion for the first three points mentioned is the internal consistency of the f/H values obtained under varied conditions. This leads to a precision of about 0.1 percent for the relative moment values of Li^6, Li^7 and F^{19}. From a consideration of the small half-widths observed for the resonance curves ($\sim$1 percent) and their symmetrical character it seems unlikely that any interactions are present which will tend to shift the minimum by more than 0.2 or 0.3 percent, if at all.

The simple model which we have used to discuss the principles of the method is, no doubt, insufficient to describe the finer details of the results, such as the width and shape of the resonance curves. For this purpose one must consider the various interactions between the nuclear spins of the different nuclei and their interactions with the rest of the molecular structure. The nature of other perturbations and the physical information which can be obtained from detailed observation of resonance minima will be discussed in another paper.

TABLE I. *Representative values of f/H for* Li^6, Li^7 *and* F^{19}.

NUCLEUS	MOLECULE	f MEGACYCLES PER SECOND	H GAUSS	$\frac{f}{H}$
Li^6	LiCl	2.127	3405	624.6
		2.127	3400	625.6
		2.155	3455	623.8
		2.155	3446	625.3
	Li_2	1.714	2742	625.0
		1.714	2744	624.7
	LiF	2.193	3506	625.5
		2.193	3501	626.5
Li^7	LiCl	5.611	3399	1651
		5.610	3400	1650
		6.587	3992	1650
		2.113	1278	1654
		5.552	3383	1651
	LiF	5.621	3401	1653
		6.580	3981	1653
		3.517	2133	1649
	Li_2	3.056	1862	1651
		3.084	1879	1652
		3.129	1907	1651
F^{19}	NaF	5.634	1407	4001
		5.634	1409	3998
		7.799	1949	4001
		7.799	1953	3992
		7.799	1952	3995
	LiF	4.204	1053	3994
		4.204	1055	3986

TABLE II. *Nuclear g's and magnetic moments.*

NUCLEUS	g	SPIN	MOMENT
$_3Li^6$	0.820	1	0.820
$_3Li^7$	2.167	$\frac{3}{2}$	3.250
$_9F^{19}$	5.243	$\frac{1}{2}$	2.622

DISCUSSION

One of the important objects of nuclear moment investigations is to ascertain whether the hyperfine structure of atomic energy levels can be accounted for entirely by the assumption that the nucleus interacts with the external electrons as a small magnet. The effects arising from the finite size of the nucleus and its charge distribution (isotope effect and electric quadrupole moment effect) modify slightly the h.f.s. predicted from this simple assumption but are still within the range of electromagnetic interactions. There may possibly be some other interactions with the electron which are not electromagnetic in nature but more like spin dependent nuclear forces. To this end a comparison of the ratio of the magnetic moments of two isotopes measured by our direct methods with that obtained from the results of h.f.s. measurements on the same isotopes is of interest. Since the electronic wave functions are the same for two isotopes, the ratio of the h.f.s. separations $(\Delta\nu)_1/(\Delta\nu)_2$ of a given atomic energy state should yield the ratio of the moments, μ_1/μ_2, very accurately if no other effect enters. It is to be expected that a discrepancy between these two moment ratios will be very small because of the short time which an electron spends in the region very close to the nucleus.

The ratio of the moments of the lithium isotopes, μ_7/μ_6, found by Manley and Millman[10] by the atomic beam zero moment method of measuring h.f.s. separations is 3.89. Our value is 3.963, which is about two percent higher. It is difficult to be certain, at the present time, that this difference represents a real physical

effect rather than an experimental error. Although our value can hardly be off by as much as 0.3 percent, the value given by Manley and Millman may possibly be in error because the Li^6 zero moment peak was not completely resolved from the Li^7 background. Further work along this line is clearly desirable. Another method of studying this question is through very accurate calculations of atomic wave functions (particularly in the case of Li) from which the nuclear moment can be calculated from h.f.s. data to a precision comparable with that of our direct measurements. The present status of this side of the problem is that our value of 3.250 for Li^7 is to be compared with 3.29 obtained from the measurements of Granath[14] on the h.f.s. of Li II, and the calculations of Breit and Doerman.[15] Fox and Rabi[11] find 3.14[16] from atomic beam experiments on Li I and the theory of Goudsmit[17] and that of Fermi and Segrè,[18] while Bartlett, Gibbons and Watson[19] calculate 3.33 from the same data. These differences, though small, may be significant; however, they are, as yet, within the range of accuracy claimed by the calculations.

For $_9F^{19}$, Brown and Bartlett[20] calculate values ranging from 1.9 to 3.8 from the h.f.s. data of Campbell[21] which are to be compared with our value of 2.622. For a discussion of the accuracy and reliability of these calculations see the conclusions of the papers by Bartlett and his co-workers.

From the standpoint of current nuclear theory our results for the ratio of the moments μ_{Li^7}/μ_{Li^6} diverge even more widely from the calculations of Rose and Bethe,[1] than did the previous results of Manley and Millman. In a recent paper, Bethe[22] has sought to improve the previous calculation by the use of an α-particle model. The agreement with experiment is more satisfactory than for the previous theory of Rose and Bethe. Whether this result is accidental remains to be seen from future calculations of other moments with a similar model.

This research has been aided by a grant from the Research Corporation.

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