Measurement of Nuclear Spin by the Method of Molecular Beams

The Nuclear Spin of Sodium

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A simple method of velocity selection of arbitrary resolving power for atomic and molecular beams is described. The method consists in spreading the beam into a velocity spectrum, through the action of external fields, and then selecting a portion thereof by means of a movable selector slit. Since all the atoms of the beam of the given velocity interval are utilized, this type of selection has the maximum efficiency. In addition a method of focussing the selected beam to increase intensity and resolution is applied. A ballistic method of using the surface ionization detector was evolved extending its use to beams of exceedingly low intensity. A selected beam of slow Na atoms, obtained and measured in this fashion, was analyzed in a weak and inhomogeneous magnetic field. Four distinct peaks of equal intensity were obtained which represent the $(2i+1)$ nuclear magnetic levels. The spin of the Na nucleus is accordingly equal to 3/2, in units of $h/2\pi$.

INTRODUCTION

THE most direct demonstration of the space quantization of angular momentum was the celebrated Stern-Gerlach experiment. The purpose of the experiments to be described is to demonstrate and measure the angular momentum of the nucleus directly by means of its effect on the space quantization of the angular momentum of the extranuclear configuration in a magnetic field.

The influence of a magnetic field on the different magnetic levels of an atom with nuclear spin "i" situated therein has been described in previous papers.[1,2,3] For the particular case of the alkalis in their normal $^2S_{\frac{1}{2}}$ states, values of the magnetic moments of the various magnetic levels as functions of the magnetic field are shown in Fig. 1 for the nuclear spin 3/2. The total number of magnetic levels is always $2(2i+1)$.

The schematic arrangement of the experiment is shown in Fig. 2. A narrow beam of sodium atoms, defined by slits S_1 and S_2 is allowed to pass through a magnetic velocity selector consisting essentially of an inhomogeneous magnetic field which spreads the beam into the usual velocity spectrum, the field being of sufficient intensity to cause a rather complete decoupling between electronic and nuclear spins. For sodium a field of 2000 gauss is sufficient. In this case the magnetic moments of the $2i+1$ levels arising from a particular value of m_j differ very slightly. By means of a movable selector slit S_3 a portion of the beam is selected for further analysis. After passing through S_3 the beam is homogeneous to an extent to be described below and contains atoms in equal numbers in all the magnetic levels arising from the $2i+1$ values of m_i but associated with only one value of m_j, namely, $+1/2$ or $-1/2$, depending on the direction of the field. The beam is then permitted to pass through a second

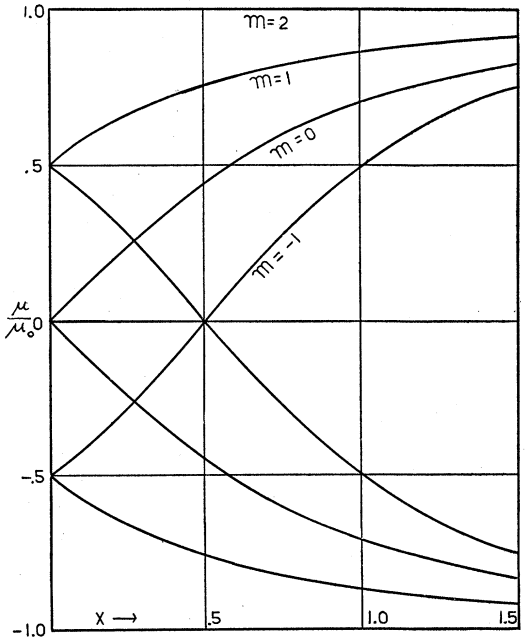


FIG. 1. Variation of the magnetic moments (in Bohr magnetons) of the atoms in the different magnetic levels for $i=3.2$ plotted against $x=(2\mu_0.hc\Delta\nu)H$.

[1] Breit and Rabi, Phys. Rev. **38**, 2082 (1931).
[2] Rabi and Cohen, Phys. Rev. **43**, 582 (1933).
[3] Rabi, Kellogg and Zacharias, Phys. Rev. **46**, 157 (1934).

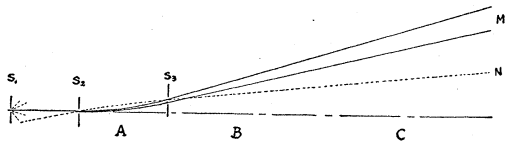


FIG. 2. Schematic arrangement of the slits and field showing the selected and scattered beams.

field of the Stern-Gerlach type. The field is so chosen that there is strong coupling between the nuclear and electronic spins. For sodium this field is of the order of 300 gauss. This brings us into the region of $x=1/2$ in Fig. 1. The atoms are subject to forces which are different for each of the $2i+1$ magnetic levels and the beam is therefore separated into $2i+1$ components, since it is homogeneous in velocity. The spin is evaluated by merely counting the number of peaks in the deflection pattern.

APPARATUS

The apparatus, (Fig. 3) constructed of solder-coated brass tubing, is divided into two sections, an oven chamber and an observation chamber. These are connected by a ground joint cemented with picien wax. Each half of the system is pumped independently, the oven chamber being connected to a fast three stage mercury diffusion pump while the observation chamber is pumped by two umbrella type single stage diffusion pumps. During the runs a "sticking" vacuum as measured by a McLeod gauge is maintained in both parts of the system. Connections for the pumps, gauge and electrical leads are made through ground joints. Mercury and grease vapors are frozen out with liquid air traps.

The oven and oven slit are constructed of nickel and show no signs of corrosion by the alkali metal. The bottom is cut with three radial grooves which rest on three nickel pegs fastened firmly to the floor of the oven chamber. This three point support permits the oven to be removed and replaced accurately to the same position. The narrowness of the pegs and the small area of contact with the oven serve to insulate it from the floor of the chamber which is water-cooled.

The oven is heated by two spirals of 10 mil tungsten wire enclosed in holes drilled in the body of the oven above and below the slit. This arrangement serves to keep the region near the slit at a temperature slightly higher than the rest of the oven. A copper constantan thermocouple is inserted into the body of the oven for temperature measurement. The oven well is covered with a tapered monel metal plug. This plug is hollowed out to permit the alkali vapor to reach the slit through a canal and at the same time to check the creeping of the liquid metal along the nickel surface to the slit. The jaws forming the oven slit are set 0.013 mm apart.

The collimating slit S_2, 0.016 mm wide and 9 cm from the oven slit, is set in a disk which can be rotated by means of a screw about the axis of the beam. If the observation chamber and one-half of the collimating slit be removed, the oven slit is observable from the observation side of the collimating slit. The one edge of the slit remaining is then adjusted parallel to the oven slit with the aid of a telemicroscope and a filar micrometer. The two slits must then be adjusted laterally so that the plane of the beam is aimed parallel to the edges of the pole peices of the magnets. This is determined by sighting at the oven slit through the observation chamber with a telescope and filar micrometer.

THE VELOCITY SELECTOR

A large Dubois magnet (A, Figs. 2 and 3) fitted with pole pieces of the Stern-Gerlach type 6 cm long with a furrow 8 mm wide and a knife edge 4 mm distant furnishes the magnetic field. The selector slit S_3, 0.035 mm wide, was mounted eccentrically on a ground joint and was movable by rotating the joint. The distance from collimator to the selector was 10 cm and the two were adjusted parallel to each other by rotation of the large ground joint which connects the oven chamber to the detecting chamber.

Since the deflection of an atom in a constant field of force perpendicular to its original direction of motion is given by $s=\text{const.}/v^2$ we have $dv/v=ds/2s$. Under the conditions of this experiment dv/v was of the order of 1/10. The velocity selected is varied simply by changing the magnetic field. The resolution can be varied by changing the position of the selector slit.

Where it is undesirable to make the velocity selection with a magnetic field, the same result

can probably be attained by spreading the beam into a velocity spectrum by means of an inhomogeneous electric field. For heavy atoms or molecules even the gravitational field of the earth is not out of the question. The principal technical difficulty which is always involved when a velocity selection is made is to detect the much enfeebled beam.

The Analyzer

The analyzer follows directly after the selector slit and consists of two magnets having pole pieces similar to magnet A but the first being 18 cm long and the second 16 cm, with 1.5 cm space between the two. The first magnet B supplies the weak analyzing field and is made long in order to obtain sufficient deflection with the small forces available. Magnet C has a strong field and serves as a focussing device. The pole pieces of the magnets are so arranged that the furrows of the B and C magnets are on the same side of the beam as the knife edge of the A magnet. This is important for the purpose of resolving the different components. The reason for this can be seen by considering the fact that the selected beam contains a finite velocity range. From any portion of the original beam the faster atoms pass through the selector slit on the side nearer the original beam and the slower atoms on the farther side since the slower atoms suffer the greater deflection. A subsequent deflection in a field similar to the first would therefore broaden the beam still further. Even if the field in the second magnet is reversed this is still the case because an atom with its magnetic moment oriented parallel to the field will be deflected toward the strong field (and *vice versa* for those opposed to the field). If the second field is reversed in direction the magnetic moment will change in direction to follow the direction of the field since the rate of change is small in the period of the Larmor precession. This is to be expected from the theorem of adiabatic transformability applied to the space quantization with respect to the field. However, by changing the pole pieces about, the stronger field is now in the opposite direction and the forces acting, being opposite to those in the first field cause the beam to become narrower instead of broader.

Magnet C has a strong field in which all the components are subject to the same force and the effect is to displace the whole deflection pattern toward the center and thus increase the resolution. Magnet C has also another very important rôle due to the fact that in addition to the selected beam some atoms scattered in the oven chamber pass through the collimator in the direction of the selector slit and form an additional beam. This is represented by the broken line in Fig. 2. These atoms have the Maxwellian distribution of velocities and are fast compared to the selected beam. This scattered beam comes nearer the center than the selected beam. Although these atoms form about one-five hundredth the total beam intensity, the velocity selection cuts down the intensity to such an extent that the scattered

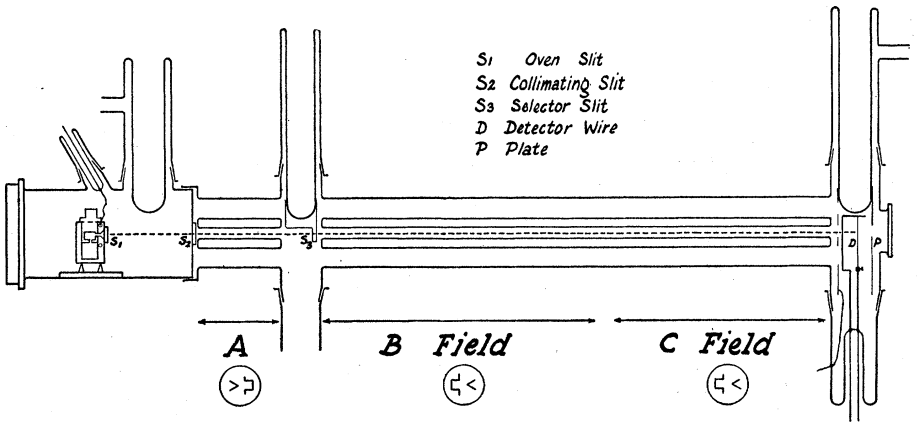


FIG. 3. Diagram of the apparatus.

atoms are as numerous as the selected atoms. The effect of magnet C is to shift the entire deflection pattern away from the region of this scattered peak so as to make the interpretation of the results unambiguous.

The Detector

Detection was effected by means of the surface ionization detector.[4] The 2 mil tungsten filament was mounted eccentrically on a ground joint and was moved laterally by turning the joint. The wire was supported by two adjustable arms so that it could be set parallel to the slit system. For the detection of sodium the wire must be provided with an oxide coat to raise its work function. No special method was used to obtain this coating. When a wire is first used it is carefully heated for a short time at about 1800°K until most of the oxide is driven off. After the beam is detected the wire is heated again until the residual positive ion current when the wire is outside the beam is reduced to about 10^{-13} amp. with the filament at a dull red heat. If too much of the oxide has been removed the wire can usually be activated by admitting a small quantity of air into the apparatus with the filament at about 1400°K. With the beam intensities used in this experiment a well sensitized filament will remain so for the entire run. The filament is maintained at a potential of 45 volts above ground. The positive ion current is measured by means of an FP-54 vacuum tube amplifier with a grid leak of 5×10^8 ohms. With the galvanometer used the current sensitivity was 10^{-14}A/mm.

The selected beam is usually too weak to measure directly because of background fluctuations. However, by allowing the sodium atoms to deposit on the cold filament from one to three minutes and then suddenly heating the filament, large ballistic throws were obtained. The multiplication factor over the direct beam readings was 20 per minute of deposition. This method eliminates the effect of background fluctuations. All the points except those of Fig. 3 were taken in this way. The galvanometer readings are almost proportional to the amount of sodium deposited if the background is less than the net ballistic reading (ballistic throws minus the background).

Procedure

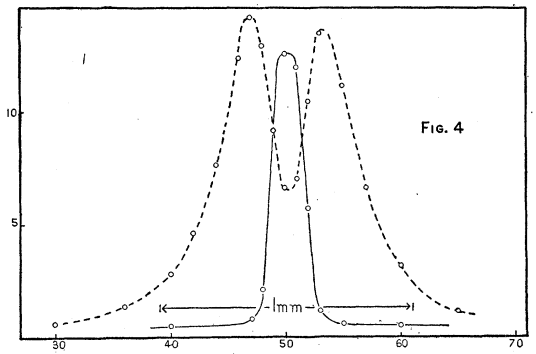
The sodium used was doubly distilled in vacuum. When the oven is to be loaded the still is cracked and the sodium dropped directly into a dish of petroleum ether and is then transferred to the oven. The lid is then pressed in, the oven set in place and the system is evacuated. During the loading process the sodium is in contact with the air for a fraction of a second and with the petroleum ether for the time taken to evacuate the oven through the slit. The oven is heated slowly to about 200°C to help remove the petroleum ether and occluded gas after which the oven is allowed to cool and stand overnight under vacuum.

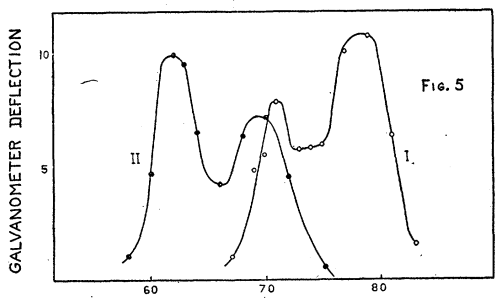
In preparation for a run the oven is heated at the rate of 3°C per minute until the beam is first detected at about 300°. The heating rate is then slowed down gradually until 360° is reached, at which temperature the beam intensity is sufficiently high for convenient working conditions. The galvanometer deflection in the center of the beam is about 10^3 cm, while the vapor pressure in the oven is about 0.2 mm Hg.

When the beam intensity has become stabilized the A field is turned on and the deflection pattern observed. The initial beam and the A field deflection pattern are shown in Fig. 4.

The A field is then thrown off and the selector slit is turned into the center of the beam. This gives a fiduciary position from which slit is turned so that atoms of the required velocity will be selected. The velocities used in this experiment correspond to an energy of about $1/2\ kT$. On Fig. 4 this would correspond to atoms which would arrive at about 80 on the scale. The low velocity is necessary in order to obtain sufficient resolution because of the small forces in the analyzing field. The resulting beam is shown in Fig. 5I. The peak at 78 is the selected beam and the one at 71 is the scattered beam. The C field is then turned on and the selected beam is deflected to the other side of the scattered beam as in Fig. 5II. It is to be noted that the scattered beam does not change its position appreciably. The analyzing field is then turned on and the deflection patterns are observed for the different values of the field.

[4] Taylor, Zeits. f. Physik **57**, 242 (1929).





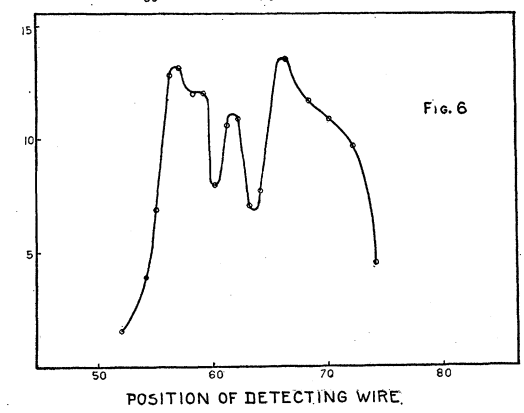
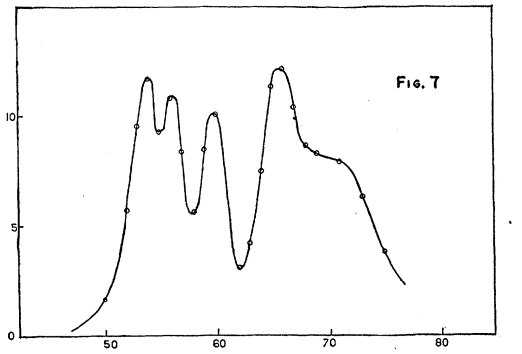
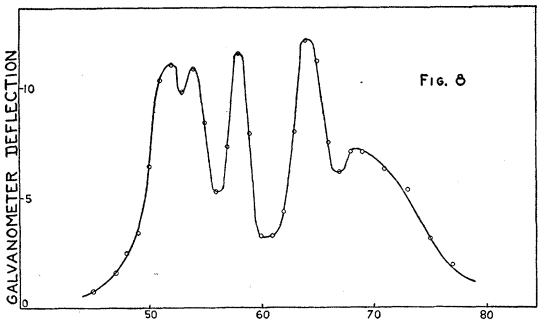


FIG. 4. Experimental curve of the direct beam (full line) and deflection pattern obtained with A field alone (dotted line). The ordinate scale of the dotted curve is 5 times that of the full curve.

FIG. 5. Experimental curve of selected and scattered beams; curve I without C field and curve II with C field. The B field was zero in each case.

FIG. 6. Deflection pattern with 55 m.a. through B magnet, A and C fields as in Fig. 5.





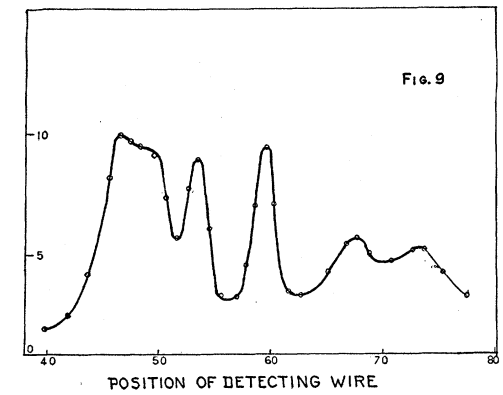


FIG. 7. Same as in Fig. 6 with B magnet current 70 m.a.
FIG. 8. Same as in Fig. 6 with B magnet current 85 m.a.
FIG. 9. Same as in Fig. 6 with B magnet current 110 m.a.

RESULTS

The different deflection patterns obtained with the various values of the current through the analyzing magnet B are shown in Figs. 6–10. In Fig. 6 the resolution is not yet sufficient to resolve all the peaks. Fig. 7, taken at a somewhat higher field shows the pattern shifted toward the left, that is, toward the position of the original beam and shows four clearly resolved peaks. The shelf on the right is due to the scattered atoms. Fig. 8 shows this still better and the scattered atoms are now almost separated from the pattern of the selected atoms. Fig. 9 shows the scattered atoms in a stronger field being split up into the ordinary Stern-Gerlach pattern, the center of which still remains at the original position of 70 scale divisions, while the deflection pattern of the selected atoms already shows the setting on of the Paschen-Back effect in the failure to resolve

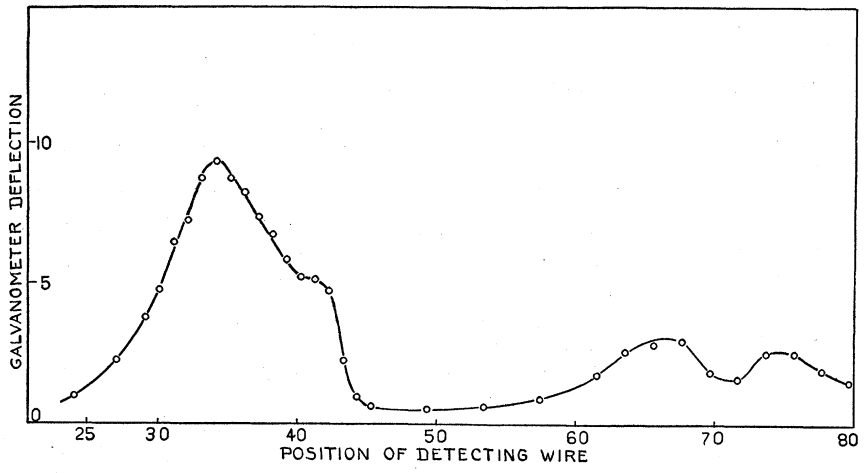


FIG. 10. Same as in Fig. 6 with B magnet current 187 m.a.

the two furthest peaks. Fig. 10, taken at a much higher field where the Paschen-Back effect is well advanced no longer shows the individual peaks. The scattered atoms are still further split in the Stern-Gerlach pattern with the center still at the same position although the selected atoms have been shifted a distance well over a millimeter. It is also of interest to note that one of the peaks in Figs. 6 and 7 are actually to the right of the position of the selected beam showing that these components have magnetic moments in weak fields opposite in sign to the strong field case as is to be expected for values of x less than $1/2$ (Fig. 1).

The four peaks obtained are of practically the same intensity, which is to be expected and have the width of the original beam. Since the number of peaks is equal to $2i+1$, the spin of the sodium nucleus is $3/2$ in units of $h/2\pi$.*

It should be mentioned that since this result is obtained only by counting the number of peaks, it is independent of any assumption as to the Maxwellian distribution of molecular velocities, or of any measurements of field, gradient or of the temperature of the oven.

* This value of the nuclear spin has been confirmed since our first publication (Phys. Rev. **43**, 582 (1933)) by Joffe from intensities in the band spectrum of the Na_2 molecule (Phys. Rev. **45**, 468 (1934)) by Granath and Van Atta from intensity measurements of the hyperfine structure of the resonance lines (Phys. Rev. **44**, 935 (1933)) and by Heydenburg and Ellett from the polarization of resonance radiation of Na in a weak magnetic field. (Phys. Rev. **44**, 326 (1933)).