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ABSTRACT

The nuclear spins of four neutron-deficient isotopes of rubidium have been determined by the atomic-beam magnetic-resonance method. The isotopes, with half lives between 4.7 hours and 80 days, are produced in quantities of  $10^{13}$  to  $10^{14}$  atoms by alpha spallation reactions in the Berkeley 60-inch cyclotron. Detection of the beam is accomplished by allowing the neutral beam to fall on a sulphur surface at room temperature, then removing the surface from the apparatus and counting with low-background high-efficiency scintillation counters which accept K x-rays accompanying K capture and internal conversion. Since all four isotopes are made in one bombardment, the identification is important (and is the subject of a separate discussion). The experimental results are:

$\text{Rb}^{81}$ ,  $I = 3/2$ ;

$\text{Rb}^{82}$ ,  $I = 5$  ;

$\text{Rb}^{83}$ ,  $I = 5/2$ ;

$\text{Rb}^{84}$ ,  $I = 2$  .

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### I INTRODUCTION

This paper presents the results of the first part of a program to measure the spins, magnetic moments, and hyperfine-structure anomalies of the long-lived ground states and long-lived excited states of the rubidium isotopes. By long-lived is meant having a half life greater than 10 minutes. Since this includes all rubidium mass numbers from 81 to 88, the hope is that information on the fine structure of the magnetic moment will be revealed.

The neutron-deficient isotopes are made by cyclotron bombardment with alpha particles on bromine. The number of atoms produced is very small, therefore a very efficient low-background counting technique has been developed for these experiments. Furthermore, for maximum isotope production the full energy (45 Mev) of the alpha beam was used. Unavoidably all four isotopes of interest are simultaneously produced. The spins are all observed in a given run but the identification becomes a significant problem. This part of the research has been greatly facilitated by the large quantity of literature on the rubidium isotopes. <sup>1-8</sup>

The isotopes under discussion are the 4.7-hr  $\text{Rb}^{81}$ , the 6.3-hr  $\text{Rb}^{82}$ , the 83-day  $\text{Rb}^{83}$ , and the 33.0-day  $\text{Rb}^{84}$ . The one largely unexpected result is the spin  $I = 5$  of  $\text{Rb}^{82}$ ; it is now of special interest to measure the spin of the 1.5-min  $\text{Rb}^{82}$ , but with present technique it seems impossible.

## II EXPERIMENTAL DETAILS

The spin of 4.7-hr  $\text{Rb}^{81}$  was first measured by use of the zero-moment technique.<sup>9</sup> When it became apparent that the spin of  $\text{Rb}^{82}$  is about 5 the apparatus was rebuilt to employ the magnetic resonance technique of Rabi as modified by Zacharias<sup>10</sup> and used by many others since for similar research. Figure 1 is a schematic diagram of the apparatus. The first-focusing, or A field, is 2.25 in. long; the refocusing field B is 21.25 in. long. The asymmetry in length gives an improvement of almost a factor of four in solid angle over a symmetrical magnet. The uniform field (C field) is 1.5 in. long; the rf transition field at the center of the C field 1 cm long. In addition to the usual surface ionization detector in the path of the collimated beam, a monitoring detector is inserted before the A magnet and out of the way of the direct beam. This detector monitors the beam continually, particularly when the normal detector is blocked by the sample-collecting button and serves to normalize the observed resonance. The sample-collecting button is inserted through a vacuum lock and intercepts the beam that would otherwise reach the detector. The detector is used to align the beam by means of the natural carrier  $\text{Rb}$  in the beam, to normalize the resonance efficiency of the apparatus by use of the natural  $\text{Rb}$ , and to calibrate the C field by use of the natural  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  frequencies. In this connection  $\text{Na}^{23}$  is always present and serves as a third point to fix the frequency scale.

A gain of 300 in intensity over the zero-moment method was both expected and observed, but this increase in intensity is marred by the presence of a background for the resonance apparatus that has no counterpart in the zero-moment machine. When the magnetic fields are off and the stop wire is considerably less than 0.1% of the full beam reaches the detector. When, however, the focusing fields ( $\sim 10,000$  gauss) and the C field ( $\sim 5$  gauss) are turned on, approximately 0.1% of the beam appears to reach the detector. This background is the present limit of sensitivity of the method and appears due to unwanted nonadiabatic transitions in the region of rapidly changing magnetic fields between the C magnet and the focusing magnets as first treated by Majorana.<sup>11</sup>

The experimental procedure is as follows: The radioisotopes are produced by alpha bombardment of a powdered  $\text{BaBr}_2$  target for from 1 to 36 hours and with cyclotron beam currents ranging from 15 to 90 microamperes. After the chemical extraction and oven preparation (to be described later) the oven, with its  $\text{RbBr}$ ,

and an excess of calcium is loaded into the apparatus via a vacuum lock. The oven is heated to between  $400^{\circ}$  and  $600^{\circ}\text{C}$ , at which point the RbBr is reduced by the calcium to free rubidium at a rate determined by surface contact and temperature. The effusion of rubidium is observed by the monitor and held approximately at a constant value during a run by slowly increasing the oven temperature. The C field is adjusted by observing transitions of the  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  carrier (as well as the unavoidable sodium). An exposure at a given radiofrequency and C-field setting is taken for periods of 3 to 30 minutes. The collecting button is then removed and a new button inserted at new frequency and field settings. The button is immediately counted so that preliminary answers are only one to two steps behind the observations. As many as 60 experimental points have been taken per run.

### III ISOTOPE PRODUCTION

The rubidium isotopes of mass numbers 81 through 84 are produced by  $(\alpha, \text{kn})$  reactions on  $\text{Br}^{79}$  and  $\text{Br}^{81}$  where  $k = 1, 2, 3,$  or  $4$ . No excitation functions exist for this set of reactions, but experimental results on near-by nuclei show quantitative agreement with the evaporation theory except for accidental effects such as competition between the ground state and its daughter for decay of the excited nucleus. A computation of the excitation function has therefore been made, using experimental Q values to obtain thresholds. The additional data used are the branching ratios for decay of the excited state as given by C. S. Wu<sup>13</sup> and W. O. Doggett.<sup>14</sup> By use of the evaporation model<sup>12</sup> the predicted cross sections are obtained as a function of the incident alpha-particle energy, and are shown in Fig. 2. These numbers are used in the next section to compare the observed intensities with the predicted intensities.

The target itself is designed to make use of the relatively high-current and high-current-density alpha beam at the Berkeley 60-inch cyclotron.  $\text{BaBr}_2$  melts at  $850^{\circ}\text{C}$  and boils at  $1850^{\circ}\text{C}$ . It is finely powdered and packed into a series of slots in a dural plate, and covered by a 1-mil dural foil which is held down by a cover plate whose beam opening is interrupted by thin ribs perpendicular to the slots. Heat dissipation is more than  $1000 \text{ watts/cm}^2$  for a 40-microampere beam. For those runs where no appreciable material was lost, the absolute agreement between experimental and theoretical production is on the order of a factor of two.



The RbBr product of the bombardment is separated from the approximately 2 grams of target material by a procedure of about 1 hour's duration. The target material is dissolved in 5 to 10 cc of water containing 1 to 15 mg of carrier RbBr. The barium is precipitated as the carbonate by a large excess of  $(\text{NH}_4)_2\text{CO}_3$  and removed by filtration. The solute is dried by heating and the  $(\text{NH}_4)\text{Br}$  is removed by sublimation at  $550^\circ\text{C}$ . The RbBr is dissolved in a few drops of water, placed in the oven, and dried. The yield has varied from 30% to 80%.

The question arises as to the choice of this method over, say, the bombardment of RbBr directly and the elimination of the chemistry. The first and least objection is that the RbBr is not as stable under bombardment as the  $\text{BaBr}_2$  and does not contain as much Br. The second objection is more fundamental, and points to one of the serious limitations of this research. With an isotope of a given half life there is available just that equivalent time to exhaust an oven charge and do an experiment. An excess of carrier or contaminant therefore limits the number of radioactive atoms available for an experiment to that fraction of the atoms that effuse through the oven during a time equal to the half life. It is for this reason that channeling an oven has very definite limits for an isotope of half life on the order of 1 hour. In these experiments the RbBr carrier could be controlled to give specific activities on the order of several hundred curies per gram, but the limitation in this direction has been the sodium contamination.

## IV BEAM DETECTION

The radioactive part of the beam is detected by collecting the neutral thermal beam on a surface, or 'button,' and then removing the surface from the apparatus to be counted. The first collection method tried<sup>15</sup> was the ionization of the beam by a surface ionization detector followed by the attraction of the ions to a surface by means of a potential difference. To summarize the results briefly it can be stated that the collection efficiency for the alkali metals is unity above 3 kilovolts on all surfaces, and is unity on some surfaces below 0.1 ev. The region between these limits gives collection efficiencies of about 2%, small but reproducible. Other methods tried were collection of the neutral beam by metal surfaces yielding generally irreproducible collection efficiencies ranging from 1% to 25% at 300°K. Platinum and tungsten, however, are reproducibly efficient with a value of 25% for chemically clean surfaces and unity for well outgassed surfaces. The method finally adopted was the use of sulphur; this has a unit collection efficiency which is unaffected by the phase and age of the surface, and no measurable loss of radioisotope has occurred over periods of several months. The success of this surface may be due to the strong chemical binding of the RbS molecule and the self-cleaning action of the surface; in air the sulphur forms a coat of sulphurous acid which is rapidly pumped off in the apparatus vacuum, leaving a clean sulphur layer. These results may not obtain for depositions involving more than a molecular layer of RbS; typical depositions in these experiments are  $\sim 10^{-3}$  of one molecular layer.

Because of the small number of decays per minute to be measured on some samples, a counting technique was needed that gave high efficiency with low background. The method finally chosen was x-ray counting with NaI scintillators. The isotopes decay primarily by K capture with a K x-ray on the order of 15 kilovolts. This x-ray is completely absorbed in a few mils of the crystal, and therefore crystals 1 mm thick were used. Furthermore, the collection geometry is very definite in this apparatus; the beam at the collector is 0.1 by 0.5 inch, and the width and length of the crystals used are only slightly larger. The total volume of the crystal is therefore only  $\sim 0.003 \text{ in.}^3$ , and yet it absorbs all the incident x-rays. The photomultiplier tubes themselves are selected 5819's or 6292's. The x-ray peak of Rb is very cleanly resolved on a differential pulse-height analyzer. The actual counting is done on the pulse-height analyzer, which accepts pulses from about 5 to 20 kev equivalent. The counting efficiency for the rubidium isotopes

is  $\sim 0.4$ , and the backgrounds for this setting are between 0.5 and 1.0 count per minute. A full description of the counting system is to be published elsewhere.

## V EXPERIMENTAL RESULTS

The only low-frequency transition that is observable with the flop-in apparatus when  $J = 1/2$  is that for which  $F = I + 1/2$  and  $m_F = -I - J \leftrightarrow m_F = -I - J + 1$ . The frequency of this transition is

$$\nu = -\frac{g_J}{2I+1} \frac{\mu_0}{h} H - \frac{2Ig_I}{2I+1} \frac{\mu_0}{h} H + \frac{2I}{(2I+1)^2} (g_I - g_J)^2 \frac{\mu_0^2}{h^2} \frac{H^2}{\Delta \nu} + O(H^3), \quad (1)$$

where  $\nu$  is the frequency of the transition,  $g_J = 2$  for the alkalis,  $\mu_0$  is the Bohr magneton,  $g_I$  is the nuclear g-factor  $\sim 1/2000$ , and  $\Delta \nu$  the hyperfine structure constant. For spin measurements the second term is always negligible within the line width of the apparatus. At sufficiently low values of the field the third term may be neglected. The procedure is to set the magnetic field at as low a value as is consistent with the resolution necessary to resolve possible lines corresponding to different spin values. The field is measured by use of the resonances of the carrier  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$ . The radiofrequency oscillator is set to the discrete frequencies of the spins of interest. This procedure alone does not uniquely determine  $I$ , for the third term above could be large enough to shift the resonance to a frequency characteristic of Zeeman transitions for a different value of  $I$ . In all cases, therefore, resonances have been observed at two or more values of magnetic field to insure that quadratic effects are small. Actually the hyperfine splittings have since been approximately determined and will be published separately when they are more precisely measured; for the present purposes it is sufficient to state that they are greater than 2500 Mc/sec and therefore have no effect in the spin determinations.

Figure 3 is a typical decay curve of the full beam taken with a 3-minute collection time with no magnetic fields and the stop wire removed. The decay is resolved into a 5.43-hour component and a component of  $\sim 70$  days. The 5.3-hour component is a mixture of 66%  $\text{Rb}^{81}$  and 34%  $\text{Rb}^{82}$  by activity, and the 70-day component is approximately 80%  $\text{Rb}^{83}$  and 20%  $\text{Rb}^{84}$  by activity. These are the fractions that are used to normalize the observed resonances. A predicted rate

is based on the cross sections of Fig. 2, on what is known of the level schemes of the isotopes, on such reducing factors as the Auger effect, on the counter efficiencies, and on the apparatus transmission.

The analysis of a typical bombardment is as follows: The predicted activity yields are  $2.38 \times 10^{11}$  counts per minute,  $0.942 \times 10^{11}$  cpm,  $6.08 \times 10^8$  cpm, and  $1.78 \times 10^8$  cpm for  $\text{Rb}^{81}$ ,  $\text{Rb}^{82}$ ,  $\text{Rb}^{83}$ , and  $\text{Rb}^{84}$  respectively. The total counting rate predicted is  $3.3 \times 10^{11}$  cpm. The total Rb activity produced is  $7.6 \times 10^{10}$  cpm. The ratio of  $\text{Rb}^{81}$  to  $\text{Rb}^{82}$  activity predicted is 2.5; the ratio observed is 1.9. The ratio of the combined  $\text{Rb}^{83}$  plus  $\text{Rb}^{84}$  activity to the combined  $\text{Rb}^{81}$  plus  $\text{Rb}^{82}$  predicted is  $2.4 \times 10^{-3}$ ; the ratio observed is  $2.7 \times 10^{-3}$ . The prediction for the activity ratio of  $\text{Rb}^{83}$  to  $\text{Rb}^{84}$  is 4 to 1. This is in only crude agreement with the observed decay. The over-all agreement is taken as satisfactory in view of the relatively rough assumptions that necessarily are part of the estimates.

The over-all results are displayed in Fig. 4. Observed intensities are plotted versus given spin settings for different buttons and different runs. Effects are observed only for  $I = 3/2$ ,  $I = 2$ ,  $I = 5/2$ , and  $I = 5$ . No effect is observed for the other spin values indicated. The experimental data as taken are not seen in this curve, since corrections have been made for the apparatus background and half life of each component, and the counting rate is normalized to the fraction of isotopic abundance in the main beam. This normalization shows the variation in intensity with spin as expected. The theoretical intensity is  $1/(2I+1)$  of the main beam. The effect of the stop wire and transition probability is to reduce the observed intensity to  $0.25/(2I+1)$  for the carrier beam; this is approximately the ratio in Fig. 1. This is itself a partial verification of the assignments. To discuss the experimental verification in detail it is convenient to discuss each isotope separately.

Rubidium-81;  $I = 3/2$ . This resonance was observed several times. Sufficient material was deposited to permit a reasonably accurate half-life determination. Figure 5 is such a decay curve, and the half life is observed to be 4.7 hours, which is the previously reported value.<sup>2</sup> For comparison the  $\text{Rb}^{82}$  decay is shown on the same figure.

Rubidium-82;  $I = 5$ . This resonance was also observed several times and at several values of the field. The primary identification was made by use of the half lives measured from the decay curves of the samples on the resonance buttons. The result of 6.3 hours (Fig. 5) agrees with the reported values.<sup>2</sup> As an example of the performance of the apparatus, two full resonance curves for this isotope are shown in Fig. 6.

Rubidium-83;  $I = 5/2$ . After a few days the  $\text{Rb}^{81}$  and  $\text{Rb}^{82}$  components decay away and the buttons can be re-counted for residual activity so as to determine the  $\text{Rb}^{83}$  and  $\text{Rb}^{84}$  spins. Figure 7 shows the decay curves of the buttons corresponding to  $I = 3/2$ ,  $I = 5/2$ , and  $I = 7/2$ , collected under experimentally similar conditions; after three days the  $I = 5/2$  button alone shows a significant counting rate. Figure 8 is the decay curve of this sample taken over several months. The measured half life is 82 days. This is to be compared with the reported 83-day half life.<sup>4</sup> The same counter was used to observe the decay of the full-beam sample. The measured half life is 76 days, showing the comparative enrichment of the sample of  $\text{Rb}^{83}$ ,  $I = 5/2$ .

Rubidium-84;  $I = 2$ . The low abundance of this isotope made this the most difficult determination. Figure 9 is the observed (uncorrected) resonance curve for  $\text{Rb}^{83}$  and  $\text{Rb}^{84}$  combined. The  $\text{Rb}^{84}$  resonance is not very far above background, and the existence of an actual peak was verified by repeating the observation several times. Furthermore, the decay curve of the resonance sample on the same counter as the  $\text{Rb}^{83}$  sample and the full-beam sample (Fig. 8) yielded a half life of 43 days. The experimental value for  $\text{Rb}^{84}$  is 33.0 days.<sup>5</sup> The difference is due to the relatively low abundance of the  $\text{Rb}^{84}$  and the background contamination of the  $\text{Rb}^{83}$ . An experimental estimate of this background subtracted from the decay of the  $I = 2$  peak yields a half life of 36 days, which is reasonable. Figure 10 is the comparison of the decays of spin-1 and spin-2 samples, showing the residual count for  $I = 2$ .

The identifications, then, are based on the following:

1. There are four isotopes reported of half life greater than 1 hour. There is a short-lived pair (~5 hours), one of which is odd A, the other even A. There is a long-lived pair (~30-80 days) one of which is odd A and the other even A. Only four resonances are observed for spin values and activities that can be assigned uniquely on this basis.

2. The chemistry, including the calcium reduction in the oven, is such that only an alkali is likely to be present in the beam at the oven temperatures.

3. The x-rays observed in counting are characteristic of a Z in the neighborhood of Rb and Br and cannot, for example, come from products of  $(\alpha, kn)$  on Ba except for accidental effects.

4. The measured half lives of the observed resonance peak samples are close to those reported in the literature.

5. The relative abundances observed in the full beam and in the resonances are approximately those predicted by current nuclear theory and the observed spin values.

## VI DISCUSSION

The spins of rubidium isotopes, which are now complete from mass 81 through 87, show an interesting parabolic form for the difference between the energy including pairing effects of the  $f_{5/2}$  and  $p_{3/2}$  proton levels. The ground state is apparently  $p_{3/2}$  for  $Rb^{81}$ ,  $Rb^{87}$ , and perhaps  $Rb^{86, 14}$  while the  $f_{5/2}$  appears to be favored for  $Rb^{83}$ ,  $Rb^{85}$ , and perhaps  $Rb^{84}$ . The occurrence of spin 2 in  $Rb^{84}$  and  $Rb^{86}$  but spin 5 in  $Rb^{82}$  also suggests that isomerism occurs in the even-mass rubidium nuclei. A projected investigation of the 20-minute  $Rb^{84m}$  will be of interest in this connection. Details of the coupling scheme of the neutron-proton system in odd-odd nuclei will be well understood only upon consideration of the nuclei of  $Z \pm 1$  as well as the nuclei of  $N \pm 1$ , hence similar investigations of the strontium and krypton isotopes are desirable.

The authors wish to express their thanks to Professor Segrè for his help in initially starting this program and to Professor Helmholtz for his help in problems associated with the decay schemes of the Rb isotopes.

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## LEGENDS

Fig. 1. Schematic diagram of the apparatus.

Fig. 2. The total cross section for production of the excited states of the neutron-deficient rubidium isotopes by  $(\alpha, kn)$  on  $\text{Br}^{79}$  and  $\text{Br}^{81}$  where  $k = 1, 2, 3, 4$ ;  $k = 1$  yields  $\text{Rb}^{84}$  and  $\text{Rb}^{82}$ ,  $k = 2$  yields  $\text{Rb}^{83}$  and  $\text{Rb}^{81}$ ,  $k = 3$  yields  $\text{Rb}^{82}$ ,  $k = 4$  yields  $\text{Rb}^{81}$ .

Fig. 3. The decay of an undeflected beam sample collected for three minutes on a sulfur surface.

Fig. 4. Activity observed on samples collected at rf frequencies appropriate to the spins indicated. The circles are proportional to the counting rates taken shortly after collection. The squares are proportional to counting rates measured several days after collection. All the counting rates are extrapolated to the end of the bombardment. The apparatus background is subtracted and the individual rates are normalized to the appropriate components in the beam. The intensities follow the ratio  $1/2I+1$ .

Fig. 5. The half-life determinations of the samples corresponding to  $I = 3/2$  and  $I = 5$ .

Fig. 6. Typical resonances for  $\text{Rb}^{82}$ .

Fig. 7. The decay of the samples corresponding to  $I = 3/2, 5/2,$  and  $7/2$  taken under similar conditions. The residual count for  $I = 7/2$  implies the spin of  $\text{Rb}^{83}$ .

Fig. 8. Decay curves for a direct beam sample compared to  $I = 5/2$  and  $I = 2$  samples taken in the same counter. The  $I = 5/2$  and  $I = 2$  yield half lives close to those for  $\text{Rb}^{83}$  and  $\text{Rb}^{84}$  respectively.

Fig. 9. Resonances of  $\text{Rb}^{83}$  and  $\text{Rb}^{84}$ . No apparatus background is subtracted.

Fig. 10. Decay curves for  $I = 1$  and  $I = 2$  samples taken under similar conditions.