

## Spins and Hyperfine Structure Separations of Radioactive Au<sup>198</sup> and Au<sup>199</sup>†

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The focusing atomic beam apparatus has been used to measure the spins and hyperfine structure separations of Au<sup>198</sup> (2.7 days) and Au<sup>199</sup> (3.15 days). The observed spins are 2 for Au<sup>198</sup> and  $\frac{3}{2}$  for Au<sup>199</sup>. Observed  $\Delta\nu$  for Au<sup>198</sup> is  $21\,800 \pm 150$  Mc/sec, assuming  $\mu_I$  to be positive, or  $22\,500 \pm 150$  Mc/sec, assuming  $\mu_I$  to be negative.  $\Delta\nu$  for Au<sup>199</sup> is  $11\,110 \pm 130$  Mc/sec, assuming  $\mu_I$  to be positive, or  $11\,180 \pm 130$  Mc/sec, assuming  $\mu_I$  to be negative. The high  $\Delta\nu$  for Au<sup>198</sup> made it necessary to study that element by observation of multiple-quantum transitions.

### I. INTRODUCTION

THE application of atomic beam techniques to radioactive atoms yields information which may be difficult to obtain by other means. Beam experiments determine the spins of nuclear states unambiguously, whereas spin assignments from  $\beta$ -decay systematics and  $\gamma$ -ray work are not necessarily unique. Furthermore, beam methods permit the determination of nuclear moments. Such measurements have been carried out on radioactive elements by other methods only in rare cases (e.g.,  $\gamma$ - $\gamma$  angular correlation in a magnetic field).

These considerations have special significance for atomic nuclei having an odd number of neutrons and an odd number of protons. Since only four such nuclei are stable, any systematic study of odd-odd behavior must be based on results obtained from elements that are unstable to  $\beta$  decay or  $K$  capture.

As part of a general program to obtain information about radioactive nuclei, we have determined the spins and magnetic moments of the ground states of Au<sup>198</sup> (2.7 days) and Au<sup>199</sup> (3.15 days). The apparatus used was the focusing atomic beam apparatus designed and built by Lemonick, Pipkin, and Hamilton<sup>1</sup> (hereafter referred to as LPH) and used by Lemonick and Pipkin<sup>2</sup> in their study of Cu<sup>64</sup> and Ag<sup>111</sup>. No detailed discussion of the equipment will be given here since it has been described in LPH. As a general source of information and bibliography on atomic beam work we refer the reader to the review article by Ramsey.<sup>3</sup>

### II. REVIEW OF THEORY

The interpretation of atomic beam resonance experiments<sup>3,4</sup> is found in the theory of the hyperfine structure

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<sup>1</sup> Lemonick, Pipkin, and Hamilton, Rev. Sci. Instr. (to be published).

<sup>2</sup> A. Lemonick and F. M. Pipkin, Phys. Rev. **95**, 1356 (1954).

<sup>3</sup> N. F. Ramsey in *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Part III.

<sup>4</sup> Kusch, Millman, and Rabi, Phys. Rev. **57**, 765 (1940).

of atoms which we review briefly here. In this paper, we will discuss only atoms for which  $J$  (the total electronic angular momentum in units of  $\hbar$ ) =  $\frac{1}{2}$ . According to the theory, in zero external field, the electronic state of the atom being studied is split into two substates (of different energy) characterized by the total angular momentum quantum numbers  $F = I + \frac{1}{2}$  and  $F = I - \frac{1}{2}$  depending on whether  $J$  and  $I$  (the nuclear spin) are mutually parallel or antiparallel. Each state labeled by a number  $F$  consists of  $2F + 1$  degenerate states which are conventionally labeled with quantum numbers  $m_F$  (hereafter called simply  $m$ ) running in integral steps from  $F$  to  $-F$ . Application of an external magnetic field removes this degeneracy. The energy of these states, as a function of external magnetic field  $H$ , is given by the Breit-Rabi relation<sup>3,5</sup>:

$$W(F, m) = -\frac{\Delta W}{2(2I+1)} - \frac{\mu_I}{I} H m \pm \frac{\Delta W}{2} \left( 1 + \frac{4m}{2I+1} x + x^2 \right)^{\frac{1}{2}} \quad (1)$$

$\Delta W$  is the separation of the two  $F$  levels at zero magnetic field. It is the difference in the energy of interaction of the nuclear magnetic moment with the electronic magnetic moment for  $I$  and  $J$  parallel and antiparallel.  $\mu_I$  is the nuclear magnetic moment in Bohr magnetons and  $x$  is a magnetic field parameter defined by

$$x = \frac{1}{\Delta W} \left( -\frac{\mu_J}{J} + \frac{\mu_I}{I} \right) H, \quad (2)$$

where  $\mu_J$  is the electronic magnetic moment in Bohr magnetons. We further recall the definition of the hyperfine structure splitting  $\Delta\nu = \Delta W/h$ .

The only dependence on  $F$  in Eq. (1) is given by the  $\pm$  sign.  $+$  or  $-$  is used depending on whether  $F = I + \frac{1}{2}$  or  $I - \frac{1}{2}$  respectively.<sup>6</sup>

<sup>5</sup> G. Breit and I. I. Rabi, Phys. Rev. **38**, 2072 (1931); Millman, Rabi, and Zacharias, Phys. Rev. **53**, 384 (1938).

<sup>6</sup> This discussion assumes that  $\mu_I$  is positive. The modifications for negative  $\mu_I$  are obvious.

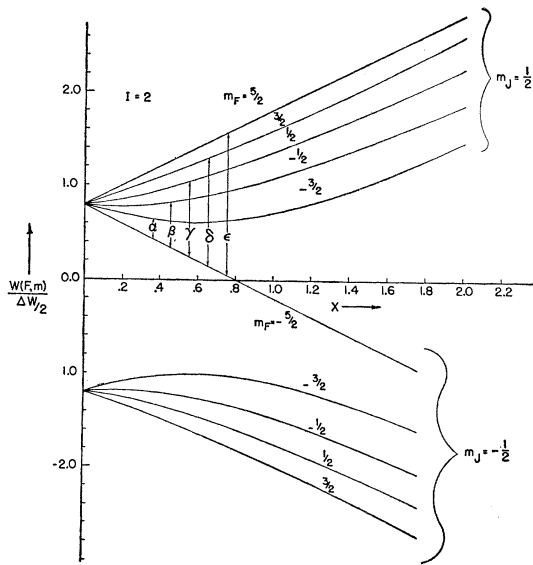


FIG. 1. Magnetic field dependence of  $W(F, m)$  [see Eq. (1)] for  $I=2$ ,  $J=\frac{1}{2}$ . In a very small field each of the states,  $F=I+J$  and  $F=I-J$ , exhibits a normal Zeeman pattern. In a very high field there are two main groups of levels corresponding to the two possible alignments of  $J$  relative to  $H$ . The separation of levels within either of the main groups at high field is due to the various possible orientations of  $I$  relative to  $J$ .

Figure 1 is a plot of Eq. (1) for  $I=2$ .<sup>7</sup> For small values of  $H$  ( $\mu_0 H/h \ll \Delta\nu$ , where  $\mu_0$  is one Bohr magneton),  $F$  is nearly a constant of the motion and for each set of magnetic sublevels labeled by a value of  $F$ , one has a normal Zeeman pattern. The Zeeman intervals at sufficiently low field are all equal and their spacing is given by

$$\nu = 1.400H/F_{\max}, \quad (3)$$

where  $\nu$  is the frequency (in Mc/sec) corresponding to the energy difference between levels  $m$  and  $m+1$  and  $H$  is given in gauss. In a very high magnetic field ( $\mu_0 H/h \gg \Delta\nu$ ) there is a so-called Back-Goudsmit region (analogous to the Paschen-Back region of fine structure theory) in which there are two main sets of levels, one corresponding to  $J$  aligned parallel to  $H$  and the other to  $J$  antiparallel to  $H$ . Here the effect of various alignments of  $I$  relative to  $J$  is to introduce a comparatively small splitting (into  $2I+1$  components) of each of the main levels,  $m_J = +\frac{1}{2}$  and  $m_J = -\frac{1}{2}$ .

### III. EXPERIMENTAL PROCEDURE

The use of atomic beams to measure nuclear quantities (when  $J=\frac{1}{2}$ ) is based on the properties of Eq. (1). We review here the application of Eq. (1) to the present experiment as described in LPH. The apparatus

<sup>7</sup> The general features illustrated by Fig. 1 and described in this paragraph are independent of  $I$ . In particular, all states having  $F=I+\frac{1}{2}$  except the one for which  $m=-(I+\frac{1}{2})$  will have positive effective magnetic moments in a high field. All states for which  $F=I-\frac{1}{2}$  as well as the one for which  $F=I+\frac{1}{2}$ ,  $m=-(I+\frac{1}{2})$  will have negative effective magnetic moments in strong field.

consists basically of: (1) an oven for producing a beam of radioactive atoms, (2) a focusing  $A$  magnet which acts on the effective magnetic moments of the atoms and transmits only those atoms for which  $m_J = +\frac{1}{2}$  (we assume for the present that atoms in the  $A$  and  $B$  fields are well out in the Back-Goudsmit region of Fig. 1) (3) a homogeneous magnetic field ( $C$  field, the intensity of which we hereafter denote by  $H_c$ ) in which the passing atoms can be subjected to electromagnetic radiation, (4) a focusing  $B$  magnet which allows only those atoms to pass for which  $m_J = -\frac{1}{2}$ , (5) a copper button upon which are deposited those atoms which have succeeded in traversing (2), (3), and (4). The number of atoms collected by the copper button can be measured by radioactive counting. From the foregoing, it will be seen that, in principle, only those atoms will get through the apparatus and be detected which have undergone a transition from a state which in a strong field has  $m_J = +\frac{1}{2}$  to one which in a strong field has  $m_J = -\frac{1}{2}$ , i.e., transitions such as  $\alpha, \beta, \gamma, \dots$  in Fig. 1. By varying the frequency of the applied radiation in the  $C$  field one observes a resonance behavior in the amount of collected radioactivity on the detector button. Curves of such resonances are shown in subsequent sections. Some atoms get through the apparatus without satisfying the conditions (2), (3), (4) and are seen as a constant background.

The value of  $H_c$ , which determines the rf frequency (for given  $F$  and  $\Delta\nu$ ) necessary to induce transitions in the element being studied, is measured by first observing the frequency required to produce the transition  $\alpha$  (Fig. 1) in  $K^{39}$  (for which  $I$  and  $\Delta\nu$  are known).

Equation (3) provides a method for nuclear spin determination.<sup>8</sup> Let  $\nu$  be the resonant frequency in the  $C$  field for atoms of unknown  $F$  and let  $\nu^{39}$  be the resonant frequency in the same field for  $K^{39}$  atoms (which are used for calibration purposes and for which  $F=2$ ). Substitution in (3) gives

$$\nu = 2\nu^{39}/F. \quad (4)$$

This determines a discrete set of frequencies (corresponding to the fact that  $F$  is integral or half-integral) at which one need search to determine  $I=F-\frac{1}{2}$ .

The condition for (4) to be valid is only that  $H$  in the  $C$  field should be low enough that both the  $K^{39}$  atoms and the atoms under investigation are in the Zeeman region.

Determination of  $\Delta\nu$  involves observing the frequency necessary to induce transitions when  $H_c$  is high enough that the atoms are no longer in the Zeeman region. Given  $\nu$ ,  $I$ , and  $H_c$  (the latter being obtained from the observation of a transition in  $K^{39}$ ) one can calculate  $\Delta\nu$  from Eq. (1).

The preceding discussion must be modified if  $\Delta\nu$  is so high that practically obtainable values of  $H$  in the  $A$  and  $B$  fields do not result in an atom's being

<sup>8</sup> Davis, Nagle, and Zacharias, Phys. Rev. **76**, 1068 (1949).

in the Back-Goudsmit region while in those fields. In this event an atom in the state ( $F=5/2$ ,  $m=-3/2$ ) in Fig. 1 may even have a negative effective magnetic moment,  $\mu_e$ , [ $\mu_e = -\partial W/\partial H$ , where  $W$  is given by Eq. (1)] in the  $A$  field, resulting in its being rejected rather than being allowed to pass through the apparatus. The transition  $\alpha$  would then be unobservable. Other transitions should, however, still be observed; e.g., the upper states of transitions  $\gamma$  and  $\delta$  in Fig. 1 always have the correct sign of  $\mu_e$  and, if the magnitude of  $\mu_e$  is great enough, these states will be focused by the  $A$  magnet. The transition  $\epsilon$  can always be observed.

To satisfy the selection rule  $\Delta m = +1$  it is necessary that all transitions shown in Fig. 1 other than  $\alpha$  proceed by means of multiple-quantum emission or absorption.<sup>9</sup> In such a transition an atom changing from a state ( $F, m$ ) to a state ( $F, m'$ ) absorbs or emits  $|m-m'|$  quanta of the applied (monochromatic) radiation; i.e., if  $\nu' = |W(F, m) - W(F, m')|/h$ , one must apply a frequency  $\nu = \nu'/|m-m'|$  to produce the corresponding multiple quantum transition. The theory<sup>10</sup> of such transitions shows that their probability is enhanced by the presence of the levels  $m''$  which lie between  $m$  and  $m'$ , and the more equally the intermediate states divide the  $m-m'$  interval, the more probable is the multiple-quantum transition. In the Zeeman region a single applied frequency, given by Eq. (3), suffices for all transitions since the energy intervals are equal; i.e., the spin determination in low field is unaffected by whether one sees only one or all of the transitions  $\alpha, \beta \cdots \epsilon$ . At fields for which the Zeeman splitting departs from normal the transitions  $\alpha, \beta \cdots$  occur at successively lower frequencies.  $\epsilon$  occurs at the lowest frequency and this frequency is given by Eq. (3)

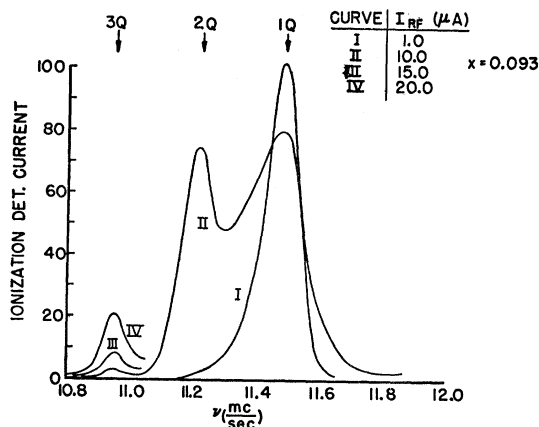


FIG. 2. One-, two-, and three-quantum transition peaks for  $K^{39}$ .  $I_{rf}$  is the current read by a microammeter in series with a germanium diode and a pickup loop which was placed near the transition-inducing hairpin loop. For these data  $I_{rf}$  was found to be proportional to the square of the rf field intensity in the transition region.

<sup>9</sup> P. Kusch, Phys. Rev. **93**, 1022 (1954), and Phys. Rev. **101**, 627 (1956).

<sup>10</sup> M. N. Hack, Phys. Rev. **100**, 975 (A) (1955).

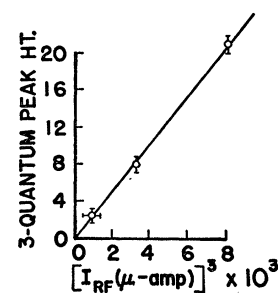


FIG. 3. Three quantum line intensity as a function of  $(I_{rf})^2$  (see Fig. 2). Hack<sup>10</sup> has predicted the linear variation shown.

regardless of the value of  $H$ . Let  $\delta\nu$  denote the difference between the applied frequency,  $\nu$ , necessary to cause an  $n$ -quantum transition and that necessary for an  $(n+1)$ -quantum transition. One can show from Eq. (1) that, for sufficiently low  $x$  (low enough that a quadratic expansion of Eq. (1) is valid),  $\delta\nu$  is independent of  $n$  and that

$$\delta\nu \Delta\nu = \nu^2. \quad (5)$$

This relation is useful for predicting where to look for multiple-quantum transitions if  $\Delta\nu$  is known. Alternatively, observation of any two adjacent multiple quantum transitions in a low field (just high enough, say, to resolve adjacent peaks) gives a rough value of  $\Delta\nu$  from Eq. (5).

Figure 2 shows some experimental multiple-quantum curves for  $K^{39}$ . The  $C$  field intensity is chosen so that the various multiple quantum peaks are resolved.  $\delta\nu$  is 0.27 Mc/sec and, using an average value of  $\nu = 11.2$  Mc/sec, one gets  $\Delta\nu$  for  $K^{39}$  equal to 465 Mc/sec which is within a percent of the accepted value<sup>3</sup> which is  $461.723 \pm 10$  Mc/sec.

The essential experimental distinction between a 1-quantum and an  $n$ -quantum transition is in the rf intensity required to produce the transition. Figure 2 illustrates the behavior of the  $K^{39}$  multiple-quantum transitions as the intensity of the applied radio-frequency signal is varied.  $I_{rf}$  is the dc current read by a microammeter which was in series with a germanium diode and a pickup loop placed in the rf field. Since this combination behaves as a square-law detector over the range of  $I_{rf}$  considered (this was verified experimentally), it follows that  $I_{rf}$  is proportional to  $H_a^2$ , where  $H_a$  is the amplitude of the applied oscillating magnetic field. Curve I is for a value of  $H_a$  such that the 1-quantum transition is starting to saturate<sup>11</sup>; no other transitions appear. The rf power applied to the atoms in curve II is 10 times that for curve I; 2 and 3 quantum transitions have appeared. Curves III and IV show the further variation of the 3-quantum peaks with successively increasing amounts of rf power. Hack<sup>10</sup> has shown that for sufficiently low  $H_a$ , the height of the  $n$ -quantum peak should vary as  $H_a^{2n}$ . Figure 3 illustrates the experimental verification of this

<sup>11</sup> We regard saturation as being that situation for which further increase of  $H_a$  results in no increase in resonance peak height.

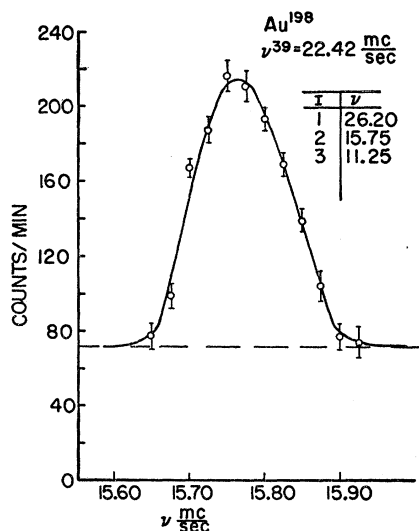


FIG. 4. One of the spin determining resonances for  $\text{Au}^{198}$ . The table shows the frequency at which a resonance would be expected to occur under various assumptions for  $I$ . Spin for  $\text{Au}^{198}$  is 2.

for the 3-quantum transition in  $\text{K}^{39}$ . It will be seen that the 3-quantum peak height varies as  $H_a^6$ .

#### IV. EXPERIMENTAL DETAILS AND RESULTS; $\text{Au}^{198}$

The  $\text{Au}^{198}$  was prepared by irradiation of natural gold in the Brookhaven Pile. One oven loading of  $\text{Au}^{198}$  consisted of  $\sim 7.5$  millicuries. The molybdenum oven described in LPH was heated to  $\sim 1150^\circ\text{C}$  as determined by observing it with an optical pyrometer. This oven temperature gave a counting rate background of 50–100 counts/min for a five-minute exposure of the collecting buttons. Of this background, about 20 counts/min were present when no exposed button was being counted, i.e., counts due to cosmic rays and naturally occurring radioactivity in the counter housing. Rather than the Geiger counter described in LPH, the counter used in this experiment consisted of a  $2\frac{1}{2}$ -in. diameter, 1-mm thick piece of trans-stilbene mounted in a Lucite cylinder and viewed by a Dumont 3-in. photomultiplier tube (type 6363). The pulses from the photomultiplier went to a conventional amplifier and scaler.

Observation of resonances at low values of  $H_c$  showed the spin of  $\text{Au}^{198}$  to be 2. Figure 4 shows one of these resonances. In this particular case  $H_c$  was high enough that Eq. (3) was not applicable to the  $\text{K}^{39}$  resonance.  $H_c$  was therefore calculated from  $\nu^{39}$  and Eq. (1). Equation (3) did apply, however, to the  $\text{Au}^{198}$  resonance.

Attempts to measure  $\Delta\nu$  by observing resonances at higher values of  $H_c$  (static  $C$  field intensity) were at first unsuccessful. Application of amounts of rf power comparable to those which had been sufficient to give strong resonance peaks in  $\text{Cu}^{64}$  and  $\text{Ag}^{111}$  in the experiment of Lemonick and Pipkin,<sup>2</sup> gave no observable

transitions in  $\text{Au}^{198}$ . The possibility was recognized that  $\Delta\nu$  might be very high thus making the ordinarily observed one-quantum transition,  $\alpha$ , unobservable (see Sec. III). In this case the spin determining resonances would have consisted only of some or all of the transitions  $\beta, \gamma, \delta, \epsilon$ . This would explain the disappearance of the resonance for high  $H_c$  since the amount of power necessary for a given multiple-quantum transition goes up rapidly as  $x$  increases.<sup>10</sup>

To determine whether the situation described above existed, a higher rf field intensity was applied<sup>12</sup> in the  $C$  region. The resonance shown in Fig. 5(a) was then observed. Subsequent runs with higher values of  $H_c$  yielded the resonance curves of Figs. 5(b) and 5(c).

If one neglects  $\mu_I$  in Eq. (1) and expands the radical keeping only terms up to order  $x^2$ , the frequency for a transition between levels  $m$  and  $m'$  (see Sec. III)

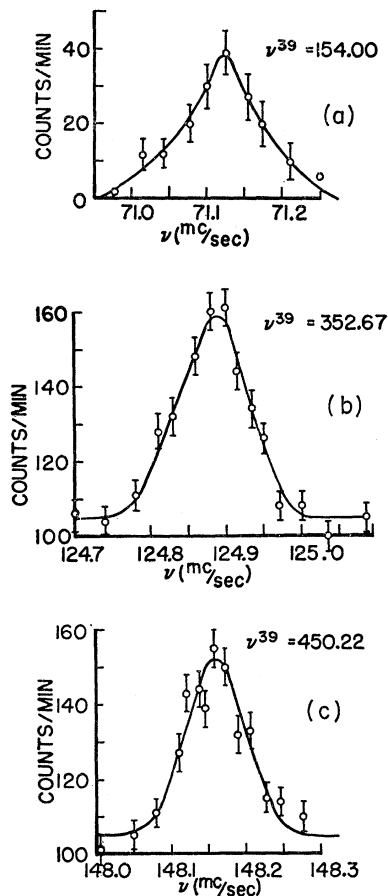


FIG. 5. Set of  $\Delta\nu$  determining resonances for  $\text{Au}^{198}$ . These resonances do not determine  $\Delta\nu$  uniquely since  $n$ , the order of the multiple quantum transition involved, is not yet known. The high rf intensity necessary to give these peaks makes it unlikely that  $n=1$ .

<sup>12</sup> A surplus jammer type AN/APT-2 was used. Hewlett-Packard oscillators type 608A and 650A were ordinarily used in observing one-quantum transitions.

becomes

$$\nu(mm') = 1.4 \frac{H}{F} - \left(1.4 \frac{H}{F}\right)^2 \left(\frac{m+m'}{\Delta\nu}\right), \quad (6)$$

which gives

$$\Delta\nu = \left(1.4 \frac{H}{F}\right)^2 (m+m') / \left(1.4 \frac{H}{F} - \nu\right). \quad (7)$$

Let  $H_1$  and  $\nu_1$  be the  $C$  field strength and frequency at which a resonance is observed. Substitution in Eq. (7) gives a value of  $\Delta\nu$  which can be used in Eq. (6) to determine  $\nu_2$ , the frequency at which a resonance should be observed for a field  $H_2$ :

$$\nu_2 - 1.4 \frac{H_2}{F} = \left(\frac{H_2}{H_1}\right)^2 \left(\nu_1 - 1.4 \frac{H_1}{F}\right). \quad (8)$$

In this approximation, then,  $\nu_2$  is independent of  $m$  and  $m'$ . It was found that, within experimental error, Eq. (8) was satisfied by the three resonances of Fig. 5. Thus the curves of Fig. 5 determine  $\Delta\nu$  uniquely only if one knows what  $m$  and  $m'$  are. This is illustrated by Table I which shows the calculated [from Eq. (1) neglecting  $\mu_I$  and not taking the anomalous  $g_J$  of Au or K into account] values of  $\Delta\nu$  for the data of Fig. 5 assuming the observed transition to be  $\alpha$ ,  $\beta$ , or  $\gamma$  (Fig. 1). It will be seen that any of the three assumptions for  $m$  and  $m'$  in Table I gives an equally consistent set of  $\Delta\nu$ 's from the experimental data.

It may be noted that for certain ranges of  $x$  there are other transitions than the set  $\alpha, \beta, \dots, \epsilon$  which meet the requirements necessary for a resonance (as described in Sec. III). In particular, if  $\Delta\nu$  is such that  $x$  corresponding to the  $A$  and  $B$  field strengths is low enough, transitions between any level with positive  $m$  and any level with negative  $m$  could be detected. Any transition other than one of the set  $\alpha, \dots, \epsilon$  would, however, occur at a frequency lower than  $1.4H_c/F$  and hence need not be considered in analyzing the resonances of Fig. 5 (all of which occur at frequencies greater than  $1.4H_c/F$ ). That is to say, that the observed transition *must* be one of the set  $\alpha, \dots, \epsilon$ .

To determine  $n = m - m'$ , and thus uniquely fix  $\Delta\nu$  the data shown in Fig. 6 were taken. The resonance of Fig. 5(a) was repeated and in addition a search was made at lower frequencies with successively increasing amounts of rf power applied to the  $C$  region. As shown,

TABLE I. Values of  $\Delta\nu$  from Fig. 5 on various assumptions for  $m$  and  $m'$ .

$\nu^{198}$ (Mc/sec)	$\nu^{89}$ (Mc/sec)	$\Delta\nu$ (Mc/sec) for transition ( $m \leftrightarrow m'$ )		
		$\alpha$ ( $-3/2 \leftrightarrow -5/2$ )	$\beta$ ( $-1/2 \leftrightarrow -5/2$ )	$\gamma$ ( $1/2 \leftrightarrow -5/2$ )
71.13	154.00	26 659	19 932	13 166
124.89	352.67	27 944	20 801	13 658
148.16	450.22	28 509	21 196	13 884

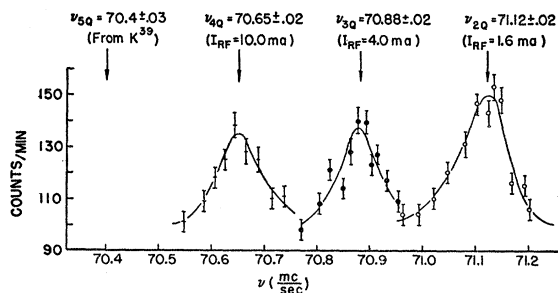


FIG. 6. Resonances observed to determine  $n$ . The  $C$  field intensity was reset to the value used in Fig. 5(a). The resonance of Fig. 5(a) was repeated and with successively increasing amounts of rf power two more resonances were observed between 71.1 and 70.4 Mc/sec. The latter frequency was calculated from the known  $C$  field as being that for which the 5-quantum transition in  $\text{Au}^{198}$  would occur. The resonances of Fig. 5 are thus identified as 2-quantum transitions.

two more resonance peaks were found between 71.12 Mc/sec and 70.4 Mc/sec. The latter frequency corresponds to the transition  $\epsilon$  in Fig. 1 and was obtained by calculation from the  $\text{K}^{39}$  resonant frequency in this same  $H_c$ . (Since this calculation was possible no attempt was made to observe the transition  $\epsilon$ ). For spin 2 this would be the frequency at which a 5-quantum transition would appear. The observed peaks of Fig. 6 are thus identified as being due to 4-, 3-, and 2-quantum transitions (compare Fig. 2). From the average spacing of the peaks of Fig. 6 one calculates, using Eq. (5),  $\Delta\nu = 20\,890$  Mc/sec which upon comparison with Table I further verifies the assignment  $n=2$  for the data of Fig. 5.

Additional verification of the multiple-quantum identification of the resonance of Fig. 6 may be found from rough data which were taken on the variation of peak height with rf intensity for each of the 3 resonances. Figure 7 is a plot of these data. The counting rate at the peak of each resonance is shown as a function of rectified rf current in the pick-up loop. The solid lines are drawn<sup>13</sup> on the assumption that  $R \propto I^n$  (which means that  $R \propto H_a^{2n}$ ) in agreement with Hack's predicted variation<sup>10</sup> of  $R$  with  $H_a$ .

The final determination of  $\Delta\nu$  based on the curves of Fig. 5 plus the knowledge that for those curves  $n=2$  is summarized in Table II. In making the calculations for Table II,  $g_J$  and  $\mu_I$  for  $\text{K}^{39}$  were taken to be 2.00228 and 0.39097 (nuclear magnetons) respectively.<sup>14,15</sup> From the work of Wessel and Lew,<sup>16</sup> on  $\text{Au}^{197}$ ,  $g_J$  for  $\text{Au}^{198}$  was taken to be 2.00412. A first approximation for  $\mu_I$  was taken to be 0.46 nm as calculated from the Fermi-Segrè formula assuming  $\Delta\nu^{198} = 20\,000$  Mc/sec and using Wessel and Lew's values of  $\Delta\nu^{197} = 6107.1 \pm 1.0$

<sup>13</sup> Note that only the slopes of these lines are significant. In each case the theoretical curve was translated to give the best fit to the corresponding data.

<sup>14</sup> Brix, Eisinger, Lew, and Wessel, Phys. Rev. **92**, 647 (1953).

<sup>15</sup> Eisinger, Bederson, and Feld, Phys. Rev. **86**, 73 (1952).

<sup>16</sup> G. Wessel and H. Lew, Phys. Rev. **92**, 641 (1953).

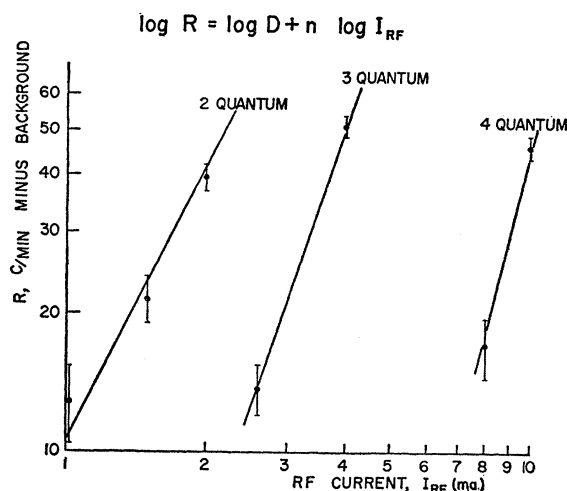


FIG. 7. Verification of the multiple-quantum order  $n$  of the resonances of Fig. 6.  $R$  is the detector counting rate taken at the peak of the various resonances. The solid lines are predicted by theory (reference 10); only their slopes are of significance.

Mc/sec and  $\mu_I^{197} = 0.13 \pm 0.01$  nm. The estimated errors of Table II are standard deviations.

The uncertainties arising in calculating  $\mu_I$  from  $\Delta\nu$  using the Fermi-Segrè formula in the case of Au (see Wessel and Lew,<sup>16</sup> Sec. V) are at present considerably greater than our experimental uncertainty in  $\Delta\nu$ . Uncertainties in the Bohr-Weisskopf correction, which is itself about 10%, can be quite appreciable. An even more important source of uncertainty comes from inability to evaluate the electronic wave functions at the nucleus accurately. Consideration of these effects has led Wessel and Lew to place an uncertainty of about 8% on their value (quoted above) of  $\mu$  for Au<sup>197</sup>. Taking either average of  $\Delta\nu$  from Table II and using the above-quoted value of  $\Delta\nu^{197}$  gives (by use of the Fermi-Segrè formula)

$$|\mu| = 0.50 \pm 0.04 \text{ nm},$$

for the magnetic moment of Au<sup>198</sup>. Here the uncertainty is taken to be the same (percentage-wise) as for Au<sup>197</sup>.

#### V. EXPERIMENTAL RESULTS; Au<sup>199</sup>

An oven loading of Au<sup>199</sup> usually consisted of 0.5 g of natural Pt which had been irradiated for a week (giving about 9 mC of activity) in the Brookhaven pile. With this much activity the oven temperature necessary to give a background counting rate of 50–100

TABLE II.  $\Delta\nu$  of Au<sup>198</sup>.

$\nu^{198}$ (Mc/sec)	$\nu^{99}$ (Mc/sec)	$\Delta\nu$ (Mc/sec) assuming $\mu_I +$	$\Delta\nu$ (Mc/sec) assuming $\mu_I -$	Estimated error
71.13	154.00	21 390	22 630	$\pm 810$
124.89	352.67	21 650	22 330	$\pm 260$
148.16	450.22	22 030	22 630	$\pm 190$
		$\Delta\nu = 21\ 800$	$\Delta\nu = 22\ 500$	$\pm 150$

counts/min for five-minute button exposures was 1200–1300°C (as read by an optical pyrometer).

In this case no difficulty was experienced in observing the transition  $\alpha$ . Observation of resonances at several values of  $H_c$  determined the spin of Au<sup>199</sup> to be 3/2 and gave the values of  $\Delta\nu$  summarized in Table III. Again, as in Table II, the estimated error of  $\pm 130$  Mc/sec for the average values of  $\Delta\nu$  is to be regarded as a standard deviation.

The calculations for Table III took into consideration the anomalous  $g_J$  for both K and Au as well as  $\mu_I$  for K<sup>39</sup> and an approximate  $\mu_I$  for Au<sup>199</sup>.

Application of the Fermi-Segrè formula assuming  $\Delta\nu^{199} = 11\ 150$  Mc/sec and referring to Sec. IV gives

$$|\mu| = 0.24 \pm 0.02 \text{ nm}.$$

for the Au<sup>199</sup> magnetic moment.

#### VI. DISCUSSION OF RESULTS

The observed spins are consistent with predictions of the nuclear shell model. According to the model<sup>17</sup> the shells being filled in the vicinity of Au are  $2d_{3/2}$  for protons and  $3p_{3/2}$  for neutrons. Thus the spin of Au<sup>199</sup>

TABLE III.  $\Delta\nu$  of Au<sup>199</sup>.

$\nu^{99}$ (Mc/sec)	$\nu^{199}$ (Mc/sec)	$\Delta\nu$ (Mc/sec) assuming $\mu_I +$	$\Delta\nu$ (Mc/sec) assuming $\mu_I -$	Estimated error
114.24	73.05	11 300	11 460	$\pm 540$
114.22	73.07	11 180	11 330	$\pm 540$
355.09	160.87	11 030	11 100	$\pm 110$
487.19	202.24	11 130	11 180	$\pm 71$
		$\text{Au} = 11\ 110$	$\text{Au} = 11\ 180$	$\pm 130$

may be regarded as due entirely to the odd  $d_{3/2}$  proton. Nordheim's "weak" rule predicts that the angular momenta of the two odd nucleons in Au<sup>198</sup> should add to a value greater than the minimum possible value of 1. The observed  $I=2$  is consistent with this rule and with the assignments  $d_{3/2}$  and  $p_{3/2}$  for proton and neutron respectively.

The observed  $|\mu|$  for Au<sup>199</sup> indicates that for this element  $\mu$  is probably positive. A negative  $\mu$  of the observed magnitude would be well outside of the lower Schmidt limit (which for  $j = \frac{3}{2} = l - \frac{1}{2}$  is  $+0.12$ ). Thus far the magnetic moments of odd- $Z$  nuclei (except the triton) have been found to be bracketed by the Schmidt lines.<sup>17</sup> Further measurements are being made to determine the signs of the moments of Au<sup>198</sup> and Au<sup>199</sup>.

The observed  $|\mu|$  of Au<sup>198</sup> is consistent with the odd group<sup>18</sup> model. Assuming the odd proton group to have the properties of Au<sup>197</sup> ( $I = \frac{3}{2}$ ;  $\mu = 0.13 \pm 0.01$ )<sup>16,15</sup> and

<sup>17</sup> M. G. Mayer and J. Hans D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley and Sons, Inc., New York, 1955), see Table VI. 1.

<sup>18</sup> Eugene Feenberg, *Shell Theory of the Nucleus* (Princeton University Press, Princeton, 1955), see Chap. III.

the odd neutron group to have the properties of  $\text{Hg}^{199}$  ( $I=\frac{1}{2}; \mu=0.499$ ),<sup>19</sup> one calculates from Feenberg's equation (III. 43)  $\mu=0.54\pm 0.01$  nm, for  $\text{Au}^{198}$ . This agrees in magnitude with the observed value. The agreement is not good, however, if one uses  $\text{Au}^{199}$  to obtain  $\mu$  for the odd proton group. In this case the calculated  $\mu$  for  $\text{Au}^{198}$  is  $0.66\pm 0.01$  nm.

Of interest in connection with  $\text{Au}^{198}$  is the work of Elliott, Preston, and Wolfson<sup>20,21</sup> who have studied the decay of  $\text{Au}^{198}$ . Their proposed disintegration scheme is shown in Fig. 8. On the basis of spin-parity assignments in  $\text{Hg}^{198}$  and the shapes and lifetimes of the  $\text{Au}^{198}$   $\beta$  spectra, Elliott *et al.* originally<sup>20</sup> concluded that the spin of the ground state of  $\text{Au}^{198}$  was 3 with odd parity. The main reasons for the assignment  $I=3$  were the shape of the 1371-keV  $\beta$  spectrum and its high  $ft$  value. Subsequent reanalysis of the data<sup>21</sup> using a more accurate calculation for determination of the shape factor results in the conclusion that the 1371-keV spectrum shape is consistent with 2- (or 3-) for the

<sup>19</sup> W. G. Proctor and F. C. Yu, Phys. Rev. **76**, 1728 (1949).

<sup>20</sup> Elliott, Preston, and Wolfson, Can. J. Phys. **32**, 153 (1954).

<sup>21</sup> Elliott, Preston, and Wolfson, Can. J. Phys. (to be published). Numerous workers have contributed to knowledge concerning the decay of this isotope. We refer the reader to reference 20 for a complete bibliography.

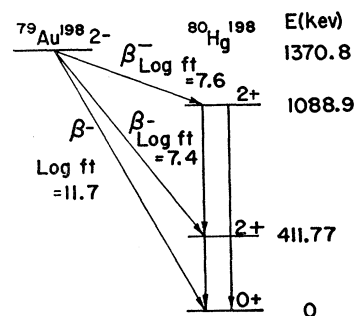


FIG. 8. Decay scheme of  $\text{Au}^{198}$  (reference 20).

$\text{Au}^{198}$  ground state, in agreement with the results of the present experiment.

Assuming for the ground state of  $\text{Au}^{198}$  an odd parity, on the basis of the  $p_{3/2}d_{3/2}$  shell model configuration assigned to it, and given the measured value of the spin reported here, the high-energy beta component is first forbidden and is about  $10^3$  times slower than typical observed transitions of this type.

#### ACKNOWLEDGMENTS

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## Scintillation Spectrometer Study of Cosmic Radiation at Great Depths

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A scintillation spectrometer has been used to observe cosmic-ray events at depths down to 10 000 feet in an oil well. The observed intensity-depth relationship can be explained in terms of the  $\mu$ -meson component and associated showers. The shower to  $\mu$ -meson ratio increases by a factor of 4 in going from sea level to a depth of 1000 meters water equivalent.

THE nature of cosmic radiation underground has been investigated in the past with the aid of G-M counter telescopes, ionization chambers, cloud chambers, and photographic emulsions.<sup>1</sup> The observations have been made in tunnels or mines and most of the experimental results are explained primarily in terms of the very penetrating  $\mu$ -meson component.

We have recently measured cosmic-ray events with a scintillation spectrometer lowered to various depths in an oil well near Houston, Texas. The spectrometer, which made use of a 2 in.  $\times$  1 $\frac{1}{2}$  in. diameter NaI(Tl) crystal, was housed in a  $\gamma$ -ray well-logging tool<sup>2</sup> and the amplified pulses from it traveled to the surface

<sup>1</sup> For a summary of underground work see E. P. George, *Progress in Cosmic Ray Physics* (North-Holland Publishing Company, Amsterdam, 1952), Chap. 7.

<sup>2</sup> McCullough Tool Company, Los Angeles and Houston.

through a 12 000-foot line. The instrument could easily be lowered to any desired depth. At the surface the pulses were again amplified, lengthened, and displayed on an Esterline-Angus recorder. The use of a pulse generator and oscilloscope permitted the calibration of the displayed pulses on the recorder in terms of the 2.62-MeV gamma ray from thorium.

For a given depth of the spectrometer the differential energy distribution of events leaving more than 9 MeV in the crystal was obtained. This level eliminates all natural gamma-ray background effects but is well below the energy ( $\sim 25$  MeV) left by a singly charged relativistic passing through the crystal. These measurements were repeated at various depths down to 10 000 feet. With a limited total time of ten days available for this experiment, it was soon evident that results