Hyperfine-Structure Separations of Au^{191} and Au^{193}†

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(Received 11 March 1964)

The hyperfine-structure separation in the $^2S_{1/2}$ electronic ground state has been measured for two radioactive gold isotopes, 3-h Au^{191} and 17.5-h Au^{193}. Atomic-beam methods, after confirming that both nuclear spins are $I=\frac{3}{2}$, have provided hfs separations of $\Delta\nu(Au^{191})=\pm5770(6)$ Mc/sec and $\Delta\nu(Au^{193})=\pm5882(10)$ Mc/sec. Combined with recent work on Au^{197}, these values give uncorrected nuclear-magnetic dipole moments of $\mu_I(Au^{191})=\pm0.136(7)$ nm, and $\mu_I(Au^{193})=\pm0.138(7)$ nm. Both measurements are consistent with the assignment of the unpaired proton in gold to a $d_{3/2}$ shell-model level.

I. INTRODUCTION

WHEN comparing experimental results with any nuclear model, it is useful to have data on several nuclei, differing among themselves by changes in neutron number. The effect of such changes in neutron number on the nuclear property being measured can then be isolated from the usually more complex problems of general nuclear structure. In this paper, we describe an atomic-beam study of the electron-nuclear hyperfine interaction of two odd-A isotopes of gold, Au^{191} and Au^{193}. The results of this study, together with similar knowledge for Au^{195}, Au^{197}, and Au^{199}, present a view of the effect on the 79th proton of additional neutron pairs between $N=112$ and $N=120$.

II. THEORY OF THE EXPERIMENT

The electronic ground state of gold is $^2S_{1/2}$. The energy of a gold atom when it is placed in a magnetic field H is thus given by the well-known Breit-Rabi equation[1] and is illustrated by Fig. 1 for a nuclear spin $I=\frac{3}{2}$. The notation is standard where $g_I=\mu_I/(\mu_0 I)$ and $g_J=\mu_J/(\mu_0 J)$ are, respectively, the nuclear and electronic g factors, μ_0 is the magnitude of the Bohr magneton, and $\Delta\nu$ is the zero-field hyperfine-structure (hfs) separation expressed as a frequency. When the magnetic field is weak the states may be distinguished by the two quantum numbers F, where $F(F+1)$ is the magnitude of the square of the vector sum of **I** and **J**, and m_F, the projection of the vector **F** along the field direction.

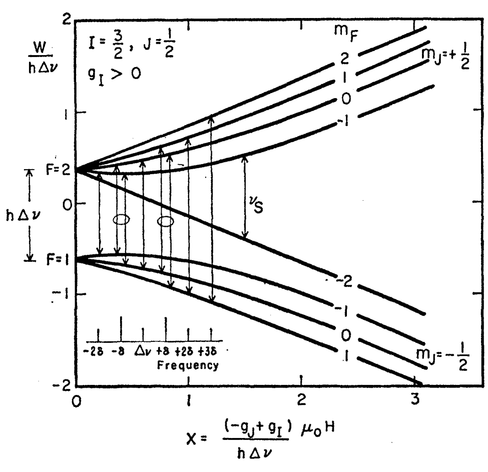

FIG. 1. Energy level diagram for a free atom with $I=\frac{3}{2}$ and $J=\frac{1}{2}$. One $\Delta F=0$ and eight $\Delta F=\pm1$ focusable transitions are shown.

A weak oscillating magnetic field can be used to mix the states, causing transitions with dipole-selection rules $\Delta m_F=0$ or $\Delta m_F=\pm1$, depending on the relative orientation of the fixed and the oscillating fields. An additional selection rule is imposed by the focusing condition of the magnetic-resonance apparatus,[2] which is able to detect only those transitions for which $\Delta m_J=\pm1$. Thus, in principle, the nine transitions shown in Fig. 1 can be observed by simple atomic-beam techniques. However, most of the resonance frequencies are approximately the magnitude of $\Delta\nu$, which usually is not known well enough to make a search practical. The $\Delta F=0$ transition marked ν_s, the so-called "standard transition," can be observed even if $\Delta\nu$ is unknown. Expanded in powers of H, the frequency of the standard transition is given by

$$\nu_s=(-g_J-2Ig_I)\frac{\mu_0}{h}\frac{H}{2I+1}+\frac{2I}{\Delta\nu}\times\left[(-g_J+g_I)\frac{\mu_0}{h}\frac{H}{2I+1}\right]^2+O\left(\frac{H^3}{\Delta\nu^2}\right). \quad (1)$$

At low fields, the resonance frequency depends on nuclear spin, but not on $\Delta\nu$. At successively higher fields, the frequency ν_s is affected by terms of higher order in H, terms that include $\Delta\nu$. Measurements of ν_s at higher fields can thus be used to determine the hfs $\Delta\nu$. The general form of Eq. (1) can be inverted quite simply to express $\Delta\nu$ as a function of ν_s:

$$\Delta\nu=\frac{[\nu_s+g_I(\mu_0/h)H][\nu_s+g_J(\mu_0/h)H]}{(-g_J-2Ig_I)(\mu_0/h)[H/(2I+1)]-\nu_s}. \quad (2)$$

† This research was supported in part by the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.
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Except for g_I, the quantities on the right are known or measureable. The value of g_I may be estimated from the Fermi-Segré formula,[3]

$$\left|\frac{g_I}{g_I'}\right|=\left|\frac{\Delta\nu}{\Delta\nu'}\right|\left(\frac{2I'+1}{2I+1}\right), \tag{3}$$

where the primed and unprimed quantities refer to different isotopes of the same element. The simultaneous solution of Eq. (2) and Eq. (3) yields consistent values of $\Delta\nu$ and g_I. However, the sign of g_I is not determined by Eq. (3). Thus, two calculations of $\Delta\nu$ must normally be made, corresponding to the assumption of a positive or negative nuclear moment. The magnetic-dipole moment of the nucleus (in nuclear magnetons) is related to the calculated g_I by

$$\mu_I(\mathrm{nm})=(M_p/m_e)g_I I, \tag{4}$$

where M_p and m_e are the proton and electron masses.

III. EXPERIMENT

The atomic-beam magnetic-resonance apparatus used for these measurements is of the flop-in design.[4] It has been described before.[5] The radioactive gold was contained in a tantalum oven heated by electron bombardment. The oven was aligned before each run with a small amount of an alkali compound that could be detected on the surface-ionization detector. Detection of the radioactive beam was accomplished by collecting samples on sulfur-coated surfaces, which were then analyzed with thin-crystal scintillation counters. The counters included a single-channel pulse-height analyzer to select only the platinum x rays following electron capture by the gold isotopes.

The details of isotope production, separation, and identification have been described previously.[6] Both Au^{191} and Au^{193} were produced by alpha bombardment of natural iridium foil, from which the gold was then evaporated by heating. Resonances of Au^{191} and Au^{193} were identified by their respective 3-h and 17-h half-lives. At lower fields, where the two resonances overlapped, a resolution was accomplished by an analysis of each resonance exposure into a 3-h and a 17-h component as described in Ref. 6.

Since the intensity of the gold atomic beam does not remain constant, a normalization has been required. The procedure used[6] is based on frequent measurements of the fast fraction of the gold beam; this fraction is only slightly affected by the deflecting fields.

The field in which the transitions took place was measured by observations of the frequency of a standard transition in rubidium. In some cases, a calibration was made with both Rb^{85} and Rb^{87}, but more frequently only Rb^{87} was used.

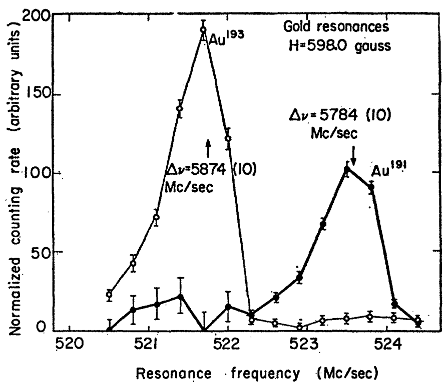

FIG. 2. Analysis of observed counting rates into 3-h Au^{191} and 17.5-h Au^{193} components.

Because of apparatus limitations, no direct $\Delta F=\pm 1$ transitions could be observed. The required frequencies (≈ 6 kMc/sec) could not be introduced successfully into the transition region. Therefore, the most precise values of hfs $\Delta\nu$ were obtained from measurements of the standard transition ν_s at the highest possible transition field, about 640 G. Typical high-field resonances are shown in Fig. 2. Here, the resonances for the two gold isotopes are just resolved.

At these high fields, the calibration frequencies were observed to drift somewhat during long experiments. The frequency of each resonance exposure was corrected for this (assumed linear) drift if any two consecutive calibrations were statistically different. Also, the time interval between calibrations was shortened so that corrections were necessary for only a few of the resonance frequencies.

The sources of rf power used to induce transitions among the Zeeman levels of the atomic system have been described previously.[7] Since the widths of the high-field $\Delta F=0$ resonances were quite large (≈ 500 kc/sec), the precision and stability of the frequency-generating and frequency-measuring equipment do not contribute to the uncertainty of the final result.

IV. RESULTS

The results of these measurements are summarized in Table I(a) for Au^{191} and Table I(b) for Au^{193}. The number in parentheses after each value represents the estimated statistical uncertainty in the least significant digit of that value. The similarity of $\Delta\nu$ results for positive- and negative-moment assumptions shows that the sign of μ_I cannot be determined from these data. The nuclear spins of Au^{191} and Au^{193} have been determined[6] as $I=\frac{3}{2}$. Therefore, from Eq. (3) the nuclear-magnetic-dipole moments will be in the same ratio as the measured hfs $\Delta\nu$'s.

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TABLE I. Gold resonances.

Resonance No.	Calibration Isotope[a]	Calibration Frequency (Mc/sec)	Magnetic field[b] (G)	Au^{191} frequency (Mc/sec)	Au^{191} $\Delta\nu$ (Mc/sec) $\mu_I>0$	Au^{191} $\Delta\nu$ (Mc/sec) $\mu_I<0$
			(a) Resonances in Au^{191}			
2281		207.90(5)	272.90(6)	211.82(10)	5711(30)	5718(30)
2311		549.29(3)	641.47(3)	571.44(20)	5758(9)	5761(9)
2421		549.94(5)	642.11(5)	571.96(10)	5766(5)	5768(5)
2422		550.00(5)	642.16(5)	571.88(15)	5772(7)	5774(7)
5661	Rb^{85} Rb^{87}	464.66(4) 505.05(3)	598.02(4)	523.60(20)	5784(10)	5786(10)
5991		530.35(10)	623.00(10)	550.80(30)	5774(14)	5776(14)
5992		530.29(10)	622.94(10)	550.70(20)	5776(10)	5778(10)
5993		530.24(10)	622.89(10)	550.65(20)	5776(10)	5778(10)
				Weighted averages:	5769(3)	5771(3)
				χ^2 of the fit (7 deg of freedom):	9.2	8.7
			(b) Resonances in Au^{193}			
Resonance No.	Calibration Isotope[a]	Calibration Frequency (Mc/sec)	Magnetic field[b] (G)	Au^{193} frequency (Mc/sec)	Au^{193} $\Delta\nu$ (Mc/sec) $\mu_I>0$	Au^{193} $\Delta\nu$ (Mc/sec) $\mu_I<0$
0693		34.31(1)	48.31(1)	34.37(15)	6895(1600)	6976(1600)
0694		59.85(2)	83.37(3)	60.07(10)	6388(370)	6424(370)
0822		104.90(8)	143.41(10)	105.70(20)	6054(240)	6071(240)
0842		169.98(2)	226.42(3)	172.10(20)	5947(84)	5957(84)
1511		204.81(2)	269.16(2)	208.10(40)	5856(110)	5863(110)
1512		204.77(2)	269.12(2)	207.93(20)	5892(57)	5988(57)
1601		437.33(10)	529.27(10)	450.00(50)	5890(30)	5893(30)
1911		433.90(3)	525.71(3)	446.44(20)	5887(14)	5890(14)
1961	Rb^{85} Rb^{87}	511.44(5) 543.84(5)	636.27(5)	562.95(20)	5878(9)	5881(9)
2241		200.46(3)	263.89(4)	203.59(20)	5865(60)	5873(60)
2281		207.90(5)	272.90(6)	211.43(10)	5815(30)	5821(31)
2311		549.98(3)	642.14(3)	569.50(50)	5873(22)	5875(22)
2831		449.36(3)	541.69(3)	462.60(40)	5894(26)	5898(26)
2832		449.38(2)	541.71(2)	462.60(20)	5896(13)	5899(13)
5661	Rb^{85} Rb^{87}	464.58(4) 504.99(3)	597.96(3)	521.75(20)	5873(10)	5876(10)
				Weighted averages:	5880(5)	5883(5)
				χ^2 of the fit (14 deg of freedom):	11.0	11.0

[a] The calibration isotope was Rb^{87} unless otherwise noted.
[b] Calculated from the calibration resonance.

The constants of the calibration isotopes Rb^{85} and Rb^{87}, as well as those of the comparison isotope Au^{197}, are listed in Table II.[8–15] These constants were used by the IBM-7090 programs HYPERFINE 4 and OMNI to calculate the hfs $\Delta\nu$ and magnetic-dipole moment for each of the radioactive-gold isotopes from each set of experimental data.

The consistency of the resonance data, as demonstrated by the values of χ^2 (Table I), is sufficient to justify the usual $\approx 70\%$ confidence in the computed uncertainties of the final averages. However, the least-squares procedure includes only statistical errors. To allow for the possibility of systematic errors in the measurements, we choose to double the purely statistical

TABLE II. Constants used in calculations.

Value	Reference
$\mu_0/h = 1.399677$ Mc/sec-G	8
$M_p/m_e = 1836.12$	8
$g_J(\mathrm{Rb}) = -2.0023457(5)$	9
$g_J(\mathrm{Au}) = -2.0033253(11)$	9
Rb^{85}: $I=\frac{5}{2}$	10
$\Delta\nu = 3035.7324$ Mc/sec	11
$\mu_I = +1.3482$ nm[a]	11
Rb^{87}: $I=\frac{3}{2}$	10
$\Delta\nu = 6834.6826$ Mc/sec	11
$\mu_I = +2.7414$ nm[a]	11, 12
Au^{197}: $I=\frac{3}{2}$	13
$\Delta\nu = 6099.320184(10)$ Mc/sec	9, 14
$\mu_I = +0.143486(9)$ nm[a]	9, 15

[a] These moments are uncorrected for diamagnetic shielding.

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errors. The best estimates for the two hyperfine-structure separations are, therefore,

$$\Delta\nu(Au^{191}) = \pm 5770(6) \text{ Mc/sec}$$

and

$$\Delta\nu(Au^{193}) = \pm 5882(10) \text{ Mc/sec.}$$

The magnetic moments can be estimated from Eq. (3), with the use of the constants for Au^{197} given in Table II:

$$\mu_I(Au^{191})_{\text{uncorr}} = \pm 0.136(7) \text{ nm}$$

and

$$\mu_I(Au^{193})_{\text{uncorr}} = \pm 0.138(7) \text{ nm.}$$

These values are not corrected for the diamagnetic shielding by the inner electrons around the nucleus, nor has the hfs anomaly been included. The hyperfine-structure anomaly (hfsa) is a measure of the inaccuracy of Eq. (3). Anomalies of more than a few tenths of a percent have not yet been observed.[16] Although no direct measurements have been made for the gold isotopes, a theoretical estimate of the hfsa can be made from the formalism of Eisinger and Jaccarino.[17] If the odd-A gold isotopes are presumed to be describable by the extreme single-particle shell model, then the anomaly between Au^{191} and Au^{197} is found to be $\approx 3\%$. An uncertainty of 5% has been adopted for the magnetic moments calculated from Eq. (3).

The diamagnetic-shielding correction[18] of 0.95% should be added before any detailed comparison with a nuclear model is made. After this correction is made we have

$$\mu_I(Au^{191})_{\text{corr}} = \pm 0.137(7) \text{ nm}$$

and

$$\mu_I(Au^{193})_{\text{corr}} = \pm 0.139(7) \text{ nm.}$$

V. DISCUSSION

The measured nuclear spins of Au^{191} and Au^{193} give further confirmation of the assignment of the 79th proton to the $2d_{3/2}$ shell-model level. A comparison of the magnetic moments of all odd-A gold isotopes is made in Fig. 3. (Strictly speaking, the comparison is of measured hyperfine-structure intervals rather than of nuclear moments, but except for the hyperfine-structure anomaly the two comparisons are equivalent. The error bars indicate the allowance for a possible 5% hfsa correction.) The magnetic moment predicted by the single-particle shell-model for a $d_{3/2}$ proton is also shown for comparison.

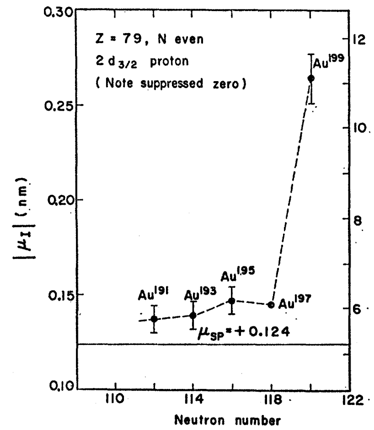

FIG. 3. The magnetic moment variation for odd-A gold isotopes with changes in neutron number.

For $N \leqslant 118$, the magnetic moments agree quite well with the single-particle value. The $\approx 10\%$ deviation from $\mu_{SP} = +0.124$ probably indicates the inadequacy of the model, while the apparent slow increase of μ_I with N may be due to hfsa corrections to the Fermi-Segré formula [Eq. (3)]. From studies of the isotope shift in odd-A isomers of mercury,[19,20] Tomlinson and Stroke have concluded that from $N = 112$ to $N = 118$, neutron pairs are added into the $i_{13/2}$ level. The present results are consistent with their conclusion since similar nuclear configurations would be expected for Au^{191}, Au^{193}, Au^{195}, and Au^{197}.

The radical change in μ_I caused by addition of the 119th and 120th neutrons indicates that the $i_{13/2}$ shell has been filled at $N = 118$. The additional pair of neutrons must then go into a new level—$f_{5/2}$, $p_{3/2}$, or $p_{1/2}$—where they may be more easily induced to contribute an effect on the magnetic moment. A measurement of the isotope shift of 44-min Hg^{199} ($I = 13/2$) would contribute greatly to the understanding of neutron levels in this region.

ACKNOWLEDGMENTS

The authors thank Dr. Y. W. Chan for his valuable contributions to the later stages of this research. The constant assistance of the staff of the late Crocker cyclotron and of the Health Chemistry group at Lawrence Radiation Laboratory is also gratefully acknowledged.

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