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# **University of California Ernest O. Lawrence Radiation Laboratory**

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HYPERFINE-STRUCTURE SEPARATIONS AND MAGNETIC MOMENTS  
OF GOLD-194, 195, AND 196

Yau Wa Chan, W. Bruce Ewbank  
William A. Nierenberg, and Howard A. Shugart

July 30, 1964

Hyperfine-Structure Separations and Magnetic Moments  
of Gold-194, 195, and 196\*

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July 30, 1964

ABSTRACT

The hyperfine-structure separations in the  $^2S_{\frac{1}{2}}$  electronic ground state of 39-h Au<sup>194</sup>, 185-d Au<sup>195</sup>, and 6.2-d Au<sup>196</sup> have been measured by the atomic-beam magnetic-resonance method. From these measurements combined with the known constants of Au<sup>197</sup>, the corresponding nuclear magnetic-dipole moments were calculated by means of the Fermi-Segrè formula. A 5% error is assigned in these moments to include a possible hyperfine-structure anomaly correction. The results are:

	$\Delta\nu$ (Mc/sec)	$\mu$ (nm)(uncorr.)
Au <sup>194</sup>	$\pm 3489.865(32)$	$\pm 0.073(4)$
Au <sup>195</sup>	$\pm 6220(38)$	$\pm 0.146(7)$
Au <sup>196</sup>	{ or    +23,100(2200) -24,700(2600)	for    +0.58(3) for    -0.62(3)

## I. INTRODUCTION

The nuclear spins of 39-h Au<sup>194</sup>, 185-d Au<sup>195</sup>, and 6.2-d Au<sup>196</sup> are known to be  $I = 1$ ,  $3/2$ , and  $2$  respectively.<sup>1,2</sup> The hyperfine-structure (hfs) separations of these isotopes in the  $^2S_{\frac{1}{2}}$  electronic ground state have now been measured by the atomic-beam magnetic-resonance technique employing the "flop-in" principle.<sup>3</sup> From the measured hfs separations, we have calculated the nuclear magnetic moments of these isotopes, using the known properties of Au<sup>197</sup> for comparison.

## II. GENERAL PRINCIPLES

For free atoms of gold in the  $^2S_{\frac{1}{2}}$  electronic ground state, the interaction of the nucleus with the valence  $s$  electron is limited to the magnetic-dipole contact interaction. This interaction splits the  $J$  level into two hyperfine levels, which are labeled by the corresponding total angular momentum  $F = I \pm \frac{1}{2}$ , where  $I$  is the nuclear spin. In the presence of an external magnetic field  $H$ , each  $F$  level splits into  $2F+1$  magnetic sublevels labeled with magnetic quantum numbers  $m_F$ . For  $J = \frac{1}{2}$ , the energy of these  $(F, m_F)$  levels is described analytically by the Breit-Rabi formula,<sup>4</sup> which may be written, in units of  $\hbar\Delta\nu$ , as

$$\frac{W(F, m_F, x)}{\hbar\Delta\nu} = -\frac{1}{2(2I+1)} - \frac{g_I}{g_I + g_J} m_F x + (F-I) \left( 1 + \frac{4m_F}{2I+1} x + x^2 \right)^{\frac{1}{2}}, \quad (1)$$

where  $x = (-g_J + g_I)(\mu_0 H / \hbar\Delta\nu)$  and the positive root is taken. Here  $g_J = \mu_J/J$  and  $g_I = \mu_I/I$  are the electronic and nuclear  $g$  factors,  $\mu_J$  and  $\mu_I$  are expressed in Bohr magnetons,  $\mu_0$  is the magnitude of the Bohr magneton, and  $\hbar\Delta\nu$  is the hfs separation energy at zero magnetic field. The value of  $g_I$

involved in this calculation is not known a priori. However, since  $\Delta\nu$  is proportional to  $g_p$ , it may be estimated, when the hyperfine structure and nuclear moment of another isotope of the same element are known, by applying the Fermi-Segrè formula<sup>5</sup>

$$\frac{|g_p|}{|g_I|} = \frac{|\Delta\nu|(2I+1)}{|\Delta\nu|(2I+1)}. \quad (2)$$

The smallness of terms in  $g_p$  ( $g_p \approx \pm g_I/2000$ ) makes it convenient to calculate the value of  $\Delta\nu$  and  $g_p$  by solving Eqs. (1) and (2) simultaneously.

Since the algebraic sign of  $g_p$  cannot be determined in this experiment, two values of  $\Delta\nu$  have been calculated--one for a positive moment and one for a negative moment. The final value of  $\Delta\nu$  is obtained as the average of these two calculations if they do not differ significantly.

A typical energy-level diagram corresponding to Eq. (1) for  $I = 2$ , and  $\mu_I > 0$  is shown in Fig. 1. The low-frequency transitions that may be observed in the atomic-beam apparatus are shown on the diagram. All these observable transitions have  $F = I + \frac{1}{2}$  and involve the level with  $m_F = -(I + \frac{1}{2})$ . A transition may be induced by absorption of  $k$  quanta of radiation, leading to a state with  $m_F = -(I + \frac{1}{2}) + k$ . A multiple quantum transition of order  $k$  (a  $k$ -quantum transition) is caused by simultaneous absorption or emission of  $k$  quanta of radiation.<sup>6</sup>

### III. PRODUCTION OF SAMPLES

Gold-194, 195, and 196 were produced at the Berkeley Crocker Laboratory's 60-in. cyclotron by the reactions  $Pt(d, kn) Au$ , for  $k = 4$  and 2. A 20-mil natural platinum foil was bombarded with 24-MeV deuterons for 12

to 24 h. A standard chemical procedure with ethyl acetate was used to separate the radioactive gold from the platinum target.<sup>7</sup> A pure metallic gold sample was obtained with about 60 mg of stable gold carrier.

In the work with Au<sup>194</sup>, the target was processed immediately after cyclotron irradiation. Because high relative specific activity of the isotope under investigation is desired for optimum results, the Au<sup>196</sup> target was stored for about 1 week (until most of the 39-h activity of Au<sup>194</sup> had decayed). A target for the study of Au<sup>195</sup> was usually stored for at least a month before use.

#### IV. EXPERIMENTAL PROCEDURE

The atomic-beam magnetic-resonance apparatus and the general procedure employed for these measurements have been discussed elsewhere,<sup>8,9</sup> so they are described only briefly here.

The gold sample was contained inside a tantalum oven lined with a carbon cup and having a carbon cap and carbon snout. Electron bombardment heated the oven to provide the beam of atoms. The radioactive beam was detected by collecting the radioactive atoms on sulfur-coated collector buttons. After exposure, each button was counted in x-ray scintillation counters containing 1 mm-thick NaI(Tl) crystals. All counting rates for each resonance button were corrected for counter background, for fluctuation of beam intensity, and for radioactive decay when necessary. The transition magnetic field was calibrated by observing the "standard transition" ( $v_g$  in Fig. 1) in Rb<sup>85</sup> and Rb<sup>87</sup> beams which were detected on a surface-ionization detector. Transition radiofrequencies were obtained with equipment described previously.<sup>9</sup> Radiofrequencies were generated and measured with enough accuracy to produce no significant error in the final results.

All data of each isotope were analyzed by computer programs to obtain the best-fit value of  $\Delta v$  according to the method of least-squares. The nuclear magnetic dipole moment of each isotope was calculated from Eq. (4) by comparison with recent measurements on Au<sup>197</sup>.<sup>10</sup> In order to allow for a possible hfs anomaly, a 5% error is assigned to magnetic dipole moments calculated from Eq. (4). The quoted moments are not corrected for diamagnetic shielding.

#### V. RESULTS FOR Au<sup>194</sup>

Several resonances of the "standard transition" in Au<sup>194</sup> have been observed at magnetic fields up to 638 G. Calculations from these resonances gave a preliminary value of  $\Delta v = 3603(40)$  Mc/sec, in agreement with the earlier measurement [ $\Delta v = 3600(120)$  Mc/sec] given by W. H. Hooke et al.<sup>11</sup> With this value, a search for  $\Delta F = \pm 1$  transitions was feasible. The experimental results of the hfs Zeeman spectrum are shown in Fig. 2. In Fig. 3, the magnetic-field-independent ( $\partial v / \partial H \approx 0$ ) transition,  $(3/2, 1/2) \leftrightarrow (1/2, -1/2)$ , was carefully observed at two different rf power levels. This test seems to indicate that the unexplained splitting of the resonance line is not due to the magnitude of the rf power.

A summary of Au<sup>194</sup> results is shown in Table I. To allow for the possibility of systematic errors in the apparatus, we take the uncertainty assigned (here and elsewhere in this paper) for the value of  $\Delta v$  to be twice that given by the least-squares analysis. The final results are  $\Delta v = \pm 3489.665(32)$  Mc/sec and  $\mu_l$  (uncorr.) =  $\pm 0.073(4)$  nm.

#### VI. RESULTS FOR Au<sup>195</sup>

Because of the long half-life of Au<sup>195</sup> (185 days), the activity produced by a cyclotron bombardment is very low. The success of producing a good

sample of Au<sup>195</sup> usually depends on how long a given Pt target can be used without being burned through by the 70-to-80- $\mu$ A deuteron beam. The practical difficulty in producing Au<sup>195</sup> samples suggested that the normal experimental procedure might be altered slightly to conserve material. The nuclear spins<sup>2,12,13</sup> of Au<sup>191</sup>, Au<sup>193</sup>, Au<sup>197</sup>, and Au<sup>199</sup> are the same as that of Au<sup>195</sup>, I = 3/2; and the corresponding hfs separations are, respectively,  $\Delta\nu \approx 5780$ , 5890,<sup>14</sup> 6099, and 11,000 Mc/sec.<sup>13</sup> It was suspected therefore, that the  $\Delta\nu$  of Au<sup>195</sup> might be some value between the  $\Delta\nu$  of Au<sup>193</sup> and Au<sup>197</sup>. By assuming that  $\Delta\nu \approx 6000$  Mc/sec., a resonance of the standard transition was first attempted and observed for Au<sup>195</sup> at a magnetic field of 273 G. A later resonance, observed at 528 G, is shown in Fig. 4. A summary of all resonances is given in Table I. The calculated values are  $\Delta\nu = \pm 6220(38)$  Mc/sec and  $\mu_L$  (uncorr.) =  $\pm 0.146(7)$  nm.

### VII. RESULTS FOR Au<sup>196</sup>

Several resonances of I = 2 with  $\Delta F = 0$  were observed at magnetic fields lower than 75 G for Au<sup>196</sup>. All these resonance frequencies show no appreciable shift from the Zeeman frequency for I = 2. This suggests that the  $\Delta\nu$  of Au<sup>196</sup> might be large. The resonance observed at a magnetic field of 84 G is shown in Fig. 5. A calculation from the small frequency shift,  $v - v_\infty \approx 0.17$  Mc/sec, indicated initially that  $\Delta\nu$  could be as large as 65,000 Mc/sec. However, in the present atomic-beam apparatus, the magnitudes of the refocusing A and B magnetic fields about 3500G; hence it is not possible to observe a standard transition for an isotope with I = 2 and  $\Delta\nu \geq 16,000$  Mc/sec. Consequently, the resonance observed could not be due to the standard transition. In this situation, the  $\Delta\nu$  of Au<sup>196</sup> may be measured either by increasing the A and B magnetic fields or by looking for multiple-quantum

transitions. Since we could not increase substantially the A and B fields, the multiple-quantum transition method was chosen. In order to observe multiple-quantum transitions, a high rf transition field is necessary in the structure (hairpin) that impresses the rf on the beam. A tunable coupling unit was connected between the hairpin and the output of the rf amplifier to improve current transfer.

The spectrum of multiple-quantum transitions terminating at the level  $m_F = - (I + \frac{1}{2})$  is labeled in Fig. 4. In Fig. 6, one of the resonance sweeps is shown. The observed resonances are identified as due to 2-quantum and 3-quantum transitions by comparing them with the corresponding transitions in Au<sup>198</sup>, which has the same spin and a similar hfs. A summary of results is given in Table I. The final calculated results are  $\Delta v = +23.4(22)$  kMc/sec with  $\mu_T(\text{uncorr.}) = +0.58(3)$  nm, or  $\Delta v = -24.7(26)$  kMc/sec with  $\mu_T(\text{uncorr.}) = -0.62(3)$  nm.

### VIII. COMMENTS

The magnetic moment of Au<sup>195</sup> is nearly the same as those of Au<sup>191</sup>, Au<sup>193</sup>, and Au<sup>197</sup> and probably arises from the 79th proton in a  $d_{3/2}$  shell.

The similarity of the spins ( $I = 2$ ) and of the magnetic moments of Au<sup>196</sup> and Au<sup>198</sup> suggests that the shell assignments and couplings in these isotopes are also the same. The magnetic moments are consistent with a ground-state configuration  $(\pi d_{3/2})(\nu p_{1/2})$  as proposed by Christensen et al.<sup>15</sup>

Removal of two neutrons from Au<sup>196</sup> ( $I = 2$ ), however, causes the ground-state spin to change to  $I = 1$ , with an accompanying change in magnetic moment. The simple shell model is not able to give a reasonable explanation for the spin  $I = 1$  for these nuclei. Although Hooke attempted to analyze the magnetic moment of odd-odd gold nuclei by using a collective model with a strongly

deformed core, the results have not been satisfactory for those nuclei with  
 $I = 4.$   $^{16}$

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FOOTNOTES AND REFERENCES

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Table I. Summary of resonances for gold-194, 195, and 196.<sup>a</sup>

Cold isotope	Calibration frequency		H (G)	Transition		$\nu_{\text{obs.}}^b$ (Mc/sec)	Frequency residual (kc/sec)	
	Rb <sup>85</sup>	Rb <sup>87</sup>		quantum numbers	$F_1 m_1$			
				$F_1$	$m_1$	$F_2$	$m_2$	
Au <sup>194</sup>	10.114(30)	15.067(30)	21.354(95)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	20.190(100) +2
	12.675(30)	18.750(30)	26.591(65)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	25.260(150) +51
	22.562(30)	33.100(30)	46.625(65)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	44.655(60) -24
	55.255(30)	78.610(30)	108.646(55)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	107.640(100) +20
	57.080(30)	81.170(10)	112.001(95)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	111.090(200) -52
	122.640(40)	164.850(50)	219.998(90)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	234.120(200) -46
	391.920(50)	-----	533.965(46)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	657.150(300) -115
	-----	545.000(100)	637.308(97)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	823.600(400) +602
	-----	545.350(50)	637.648(49)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	823.950(300) +386
	516.680(50)	548.327(50)	640.532(50)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{3}{2}$	- $\frac{3}{2}$	828.000(300) -366
	1.385(10)	-----	2.961(21)	$\frac{3}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3495.200(100) -203
	1.445(10)	-----	3.088(21)	$\frac{3}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3495.500(100) -143
	2.090(10)	3.106(10)	4.448(30)	$\frac{3}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3498.100(100) -89
	2.110(10)	3.128(10)	4.485(30)	$\frac{3}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3498.300(200) +41
	2.303(10)	3.421(10)	4.899(35)	$\frac{3}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3499.100(100) +65
	2.428(10)	3.167(10)	4.532(30)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3494.600(200) +479
	2.295(10)	3.408(10)	4.881(35)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3494.600(100) +149
	1.519(10)	2.229(10)	3.215(40)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	- $\frac{1}{2}$	3489.883(30) +8
	1.520(10)	-----	3.217(40)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	- $\frac{1}{2}$	3489.883(30) +8
	1.521(10)	2.234(10)	3.220(40)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	- $\frac{1}{2}$	3489.883(30) +8
	2.281(10)	3.387(10)	4.851(35)	$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	- $\frac{1}{2}$	3489.900(100) +12
	2.280(10)	3.398(10)	4.867(35)	$\frac{3}{2}$	- $\frac{1}{2}$	$\frac{1}{2}$	- $\frac{1}{2}$	3485.100(100) -239

Table I. (continued)

Cold isotope	Calibration frequency		H (G)	Transition		$\nu_{\text{obs.}}^{\text{b}}$ (Mc/sec)	Frequency residual (kc/sec)
	Rb <sup>85</sup>	Rb <sup>87</sup>		$F_1 m_1$	$F_2 m_2$		
Au <sup>195</sup>	159.108(50)	207.929(50)	272.886(60)	2 -1 → 2 -2	210.160(200)	+144	
	358.930(40)	411.960(20)	502.789(30)	2 -1 → 2 -2	419.250(300)	+34	
	335.919(50)	436.463(30)	528.387(40)	2 -1 → 2 -2	444.350(400)	-157	
Au <sup>196</sup>	88.288(10)	122.062(10)	165.703(20)	$\frac{5}{2} - \frac{1}{2} \rightarrow \frac{5}{2} - \frac{5}{2}$	188.000(200)	-44	
	88.288(10)	122.062(10)	165.703(20)	$\frac{5}{2} - \frac{1}{2} \rightarrow \frac{5}{2} - \frac{5}{2}$	281.010(300)	+87	
	88.277(10)	122.030(10)	165.673(20)	$\frac{5}{2} - \frac{1}{2} \rightarrow \frac{5}{2} - \frac{5}{2}$	187.950(200)	-59	
	88.277(10)	122.030(10)	165.673(20)	$\frac{5}{2} - \frac{1}{2} \rightarrow \frac{5}{2} - \frac{5}{2}$	280.875(300)	+3	
	84.915(10)	117.728(20)	160.105(25)	$\frac{5}{2} - \frac{1}{2} \rightarrow \frac{5}{2} - \frac{5}{2}$	181.600(350)	-47	
	84.915(10)	117.728(20)	160.105(25)	$\frac{5}{2} - \frac{1}{2} \rightarrow \frac{5}{2} - \frac{5}{2}$	271.530(300)	+170	

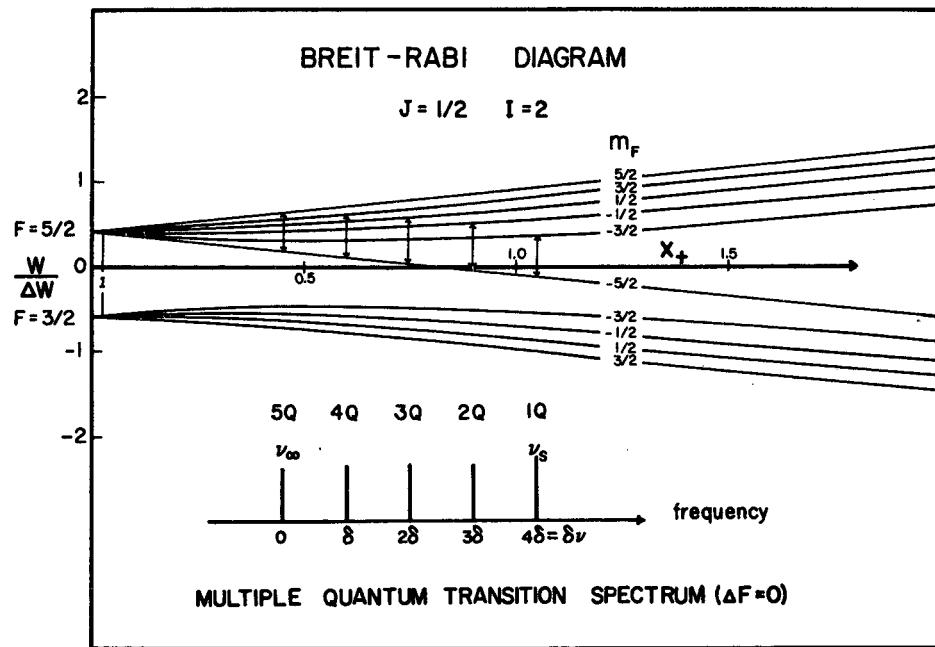
a. Results:

Isotope	I	$\Delta\nu$ (Mc/sec)	$\mu_I$ (uncorr.) (nm)	$\chi^2$	
Au <sup>194</sup>	4	±3489.865(32)	±0.073(4)	25	(22 observations)
Au <sup>195</sup>	$\frac{3}{2}$	+6218(38)	+0.146(7)	0.5	(3 observations)
	$\frac{1}{2}$	-6223(38)	-0.146(7)	0.5	
Au <sup>196</sup>	2	+23100(2200) -24700(2600)	+0.58(3) -0.62(3)	0.5 0.3	(6 observations)

b. For Au<sup>196</sup>,  $\nu_{\text{obs.}}$  is multiplied by a factor k, the order of the multiple-quantum transition.

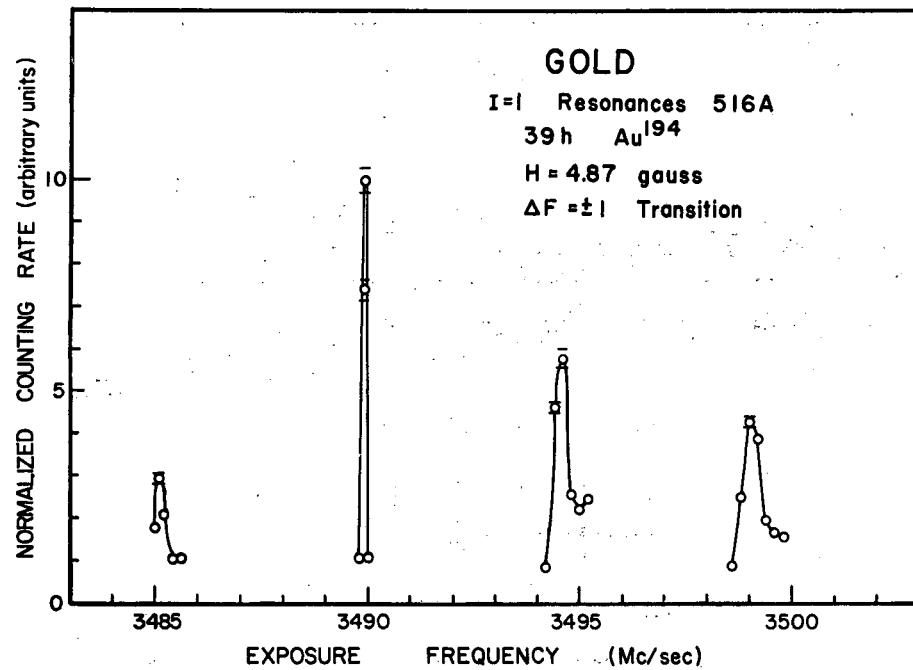
FIGURE LEGENDS

- Fig. 1. An energy-level diagram for an atom with  $J = 1/2$  and  $I = 2$ . The flop-in, multiple-quantum transitions shown all involve the  $(5/2, - 5/2)$  level.
- Fig. 2. The observed direct ( $\Delta F = \pm 1$ ) transitions in 39-h Au<sup>194</sup>.
- Fig. 3. A  $\Delta F = \pm 1$  field-independent resonance at two rf power levels.
- Fig. 4. The "standard" flop-in transition in 185-d Au<sup>195</sup>.
- Fig. 5. The  $\Delta F = 0$  transition in 6.2-d Au<sup>196</sup> at 84G.
- Fig. 6. Two- and three-quantum transitions in 6.2-d Au<sup>196</sup>.



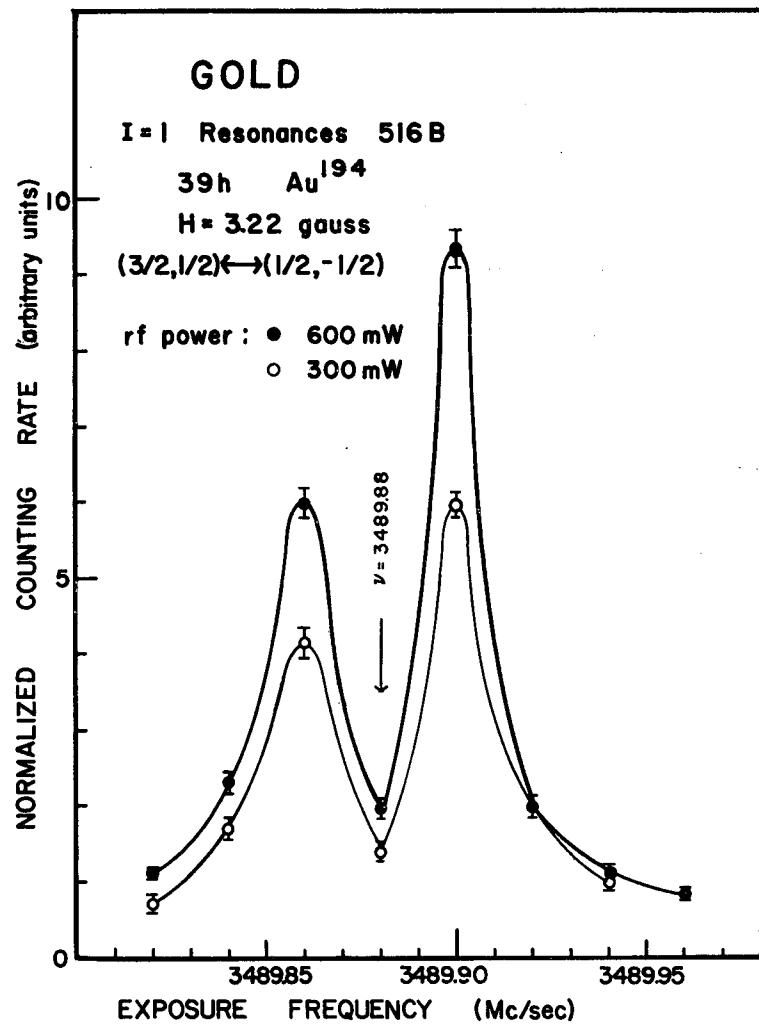
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Fig. 1



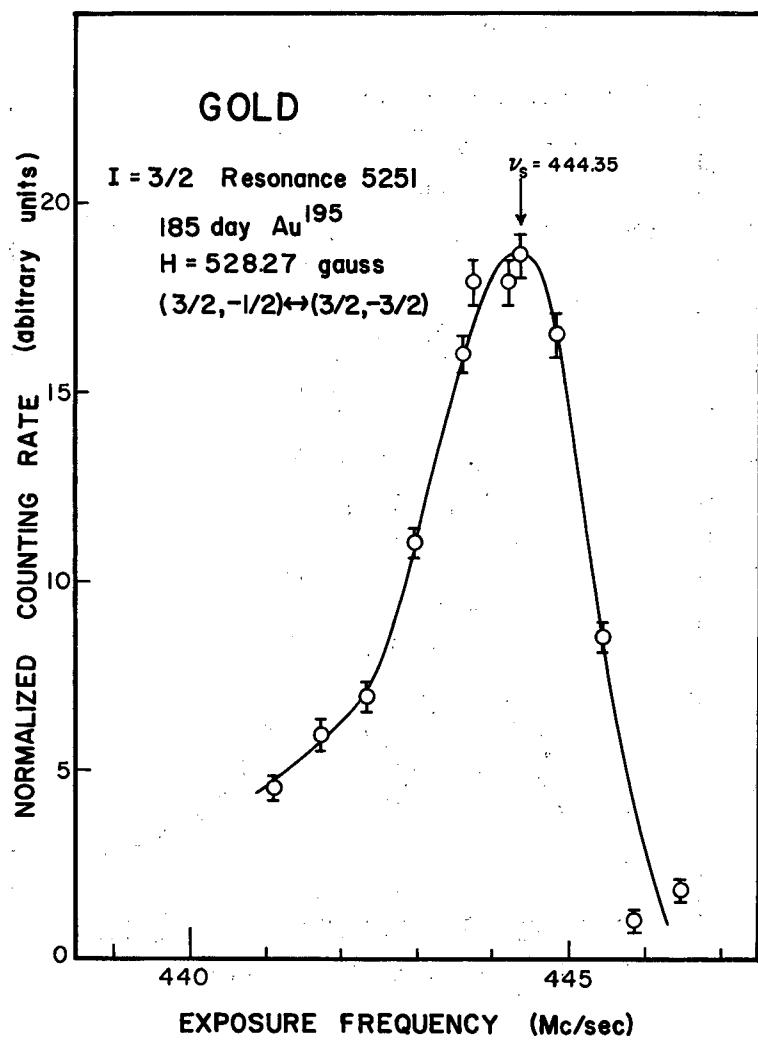
MU-27407

Fig. 2



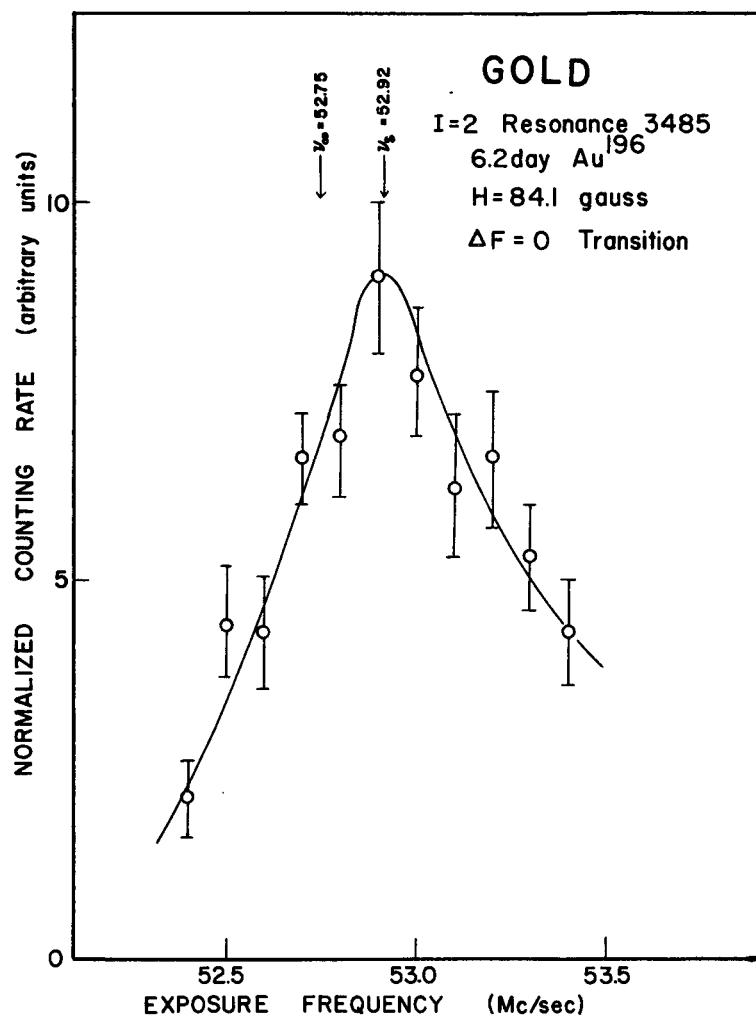
MU-27408

Fig. 3



MU-27398

Fig. 4



MU-27399

Fig. 5

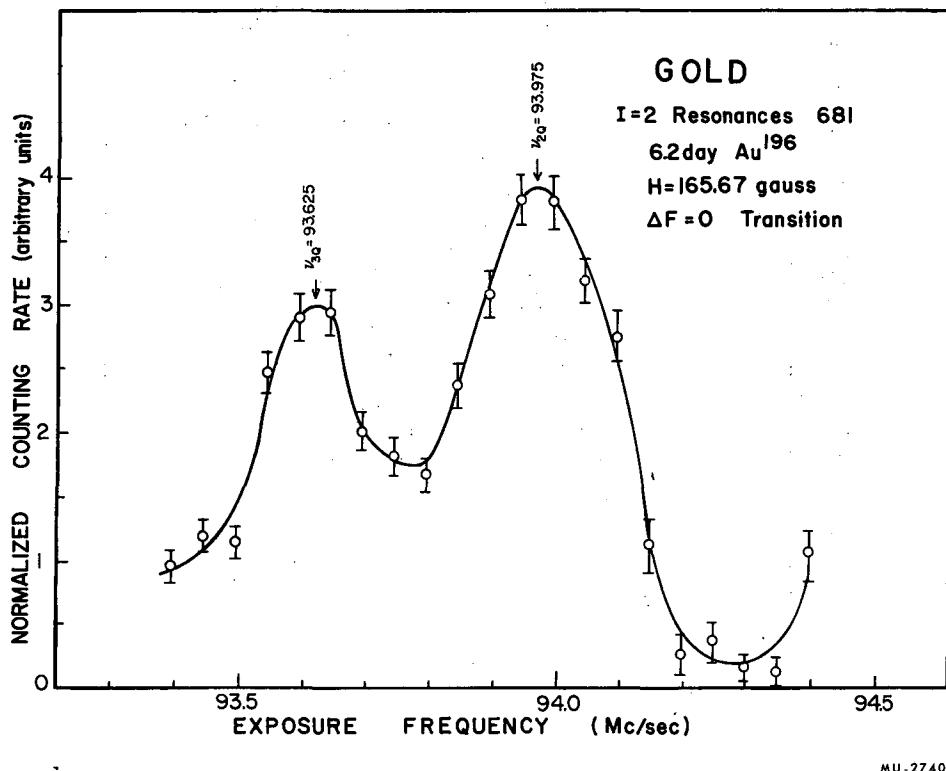


Fig. 6

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