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Determination of the Dipole Moment and Isotope Shift of Radioactive Hg¹⁹⁷ by "Double Resonance"*

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Paramagnetic resonance was established between m -sublevels ($\Delta m = \pm 1$, $\Delta F = 0$) of the 3P_1 state of radioactive Hg¹⁹⁷ at 3000 Mc/sec. From these data the nuclear interaction constant A was found to be $(513.5 \pm 1) \times 10^{-3}$ cm⁻¹, and barring hfs anomalies it lead to a ratio of moments $\mu_{197}/\mu_{199} = A_{197}/A_{199} = 1.045$; further, the nuclear spin of Hg¹⁹⁷ was ascertained to be $\frac{1}{2}$. The double resonance was combined with magneto-optic scanning to give the isotope shift of Hg¹⁹⁷, which was found to be in the 2537 Å line $+(91 \pm 5) \times 10^{-3}$ cm⁻¹ from Hg¹⁹⁸. The radioactive mercury was produced by the Au¹⁹⁷($d, 2n$)Hg¹⁹⁷ reaction and used in vapor form. Satisfactory signals were obtained with as few as 3×10^{12} atoms.

INTRODUCTION

THE hyperfine structure (hfs) of the 2537 Å line of radioactive mercury was partly analyzed in previous work by Bitter *et al.*,¹ and the dipole moment of Hg¹⁹⁷ was found to be 4% larger than that of Hg¹⁹⁹. It was decided to attempt to produce a microwave resonance at 3000 Mc/sec between the m -sublevels of the 3P_1 state of this isotope. This would provide a more accurate value (1 part in 500) for the splitting between the $F = \frac{1}{2}$ and $F = \frac{3}{2}$ levels; and a combination with magneto-optic scanning would give a reliable value for the isotope shift. Further, the feasibility of the resonance experiment at this low frequency would allow us to proceed to a 22 000-Mc/sec experiment for the direct determination of the $F = \frac{1}{2} - F = \frac{3}{2}$ interval.

The "double resonance" principle is described by Brossel and Bitter²; magneto-optic scanning, in reference 1. The combination of these two principles as

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¹ Bitter, Davis, Richter, and Young, Phys. Rev. **96**, 1531 (1954).

² J. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952).

applied to natural mercury is given by Sagalyn *et al.*³ We used the same apparatus in the present experiment and shall not redescribe the procedure and experimental arrangement. As a matter of fact, since the spins of Hg¹⁹⁷ and Hg¹⁹⁹ are both $\frac{1}{2}$, the situations are identical, so that we observed in the $F = \frac{3}{2}$ level the two resonances $m = -\frac{1}{2} \rightarrow m = -\frac{3}{2}$ and $m = +\frac{1}{2} \rightarrow m = +\frac{3}{2}$. The energy of the m -sublevels *versus* field, and the location of the resonances are shown in Fig. 1.

PREPARATION OF THE SAMPLES

Radioactive mercury was produced by bombarding a gold target with deuterons according to the Au¹⁹⁷($d, 2n$)Hg¹⁹⁷ reaction. This method for producing neutron-deficient isotopes of mercury, as well as the Au¹⁹⁷(p, xn)Hg^{198-x} reaction are well known^{4,5} with x as large as 7. The nuclear energy-level schemes are fairly well established (Fig. 2), and the odd isotopes are known to have an isomeric state because of the availability of the $i + \frac{1}{2} = 13/2$ subshell.

We used 15.2-Mev deuterons at a beam current of 40 μ amp with a 12-hour bombardment. This gives a very good yield of radioactive mercury, approximately

³ Sagalyn, Melissinos, and Bitter, Phys. Rev. **109**, 375 (1958).

⁴ Huber, Humbel, Schneider, and de-Shalit, Helv. Phys. Acta **24**, 127 (1951).

⁵ Gillon, Gopalakrishnan, de-Shalit, and Mihelich, Phys. Rev. **93**, 124 (1954).

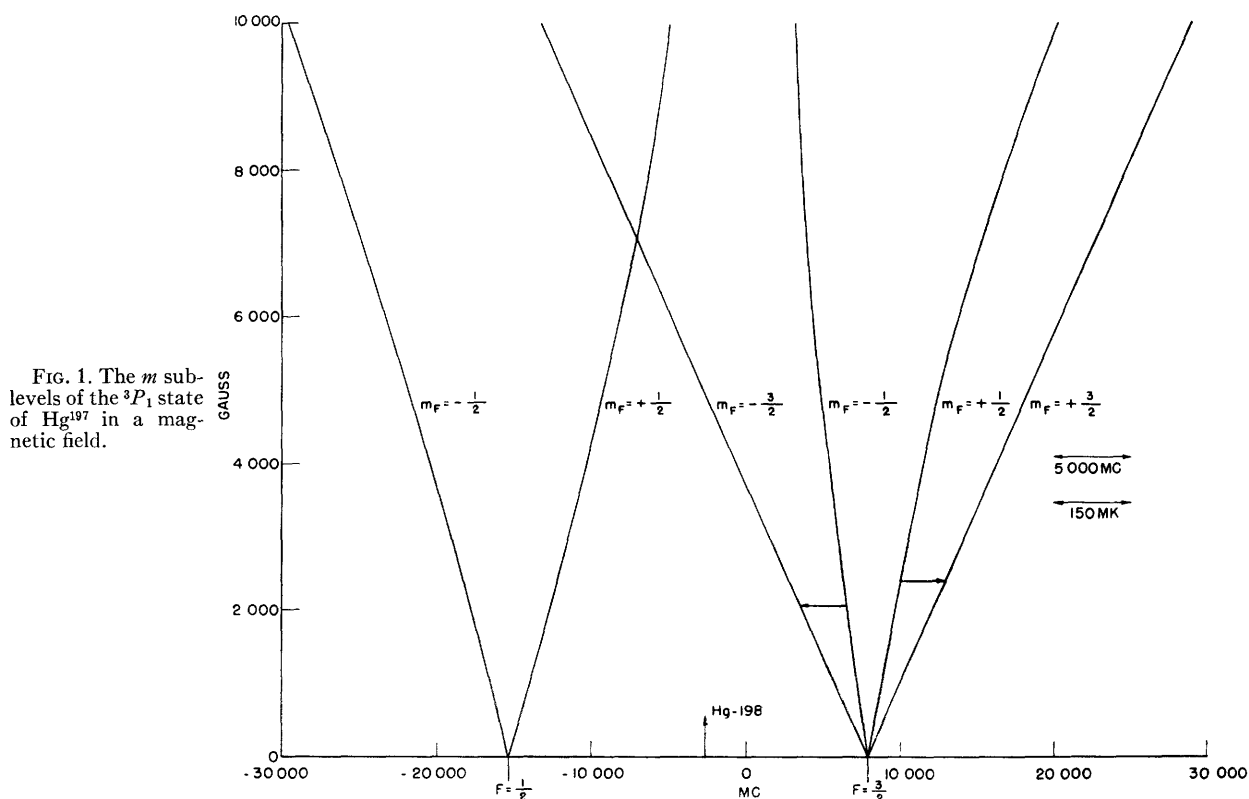


FIG. 1. The m sub-levels of the 3P_1 state of Hg¹⁹⁷ in a magnetic field.

2×10^{14} atoms. Since the threshold for the Au(d, n)Hg¹⁹⁵ reaction is above 20 Mev, Hg¹⁹⁷ and Hg^{197*} were the only radioactive isotopes produced; however, 15 Mev is well above the Au($d, 3n$)Hg¹⁹⁶ and Au(d, n)Hg¹⁹⁸ thresholds, so that stable Hg¹⁹⁶ and Hg¹⁹⁸ were produced as well. (This was determined spectroscopically.)

Since the yield was satisfactory, our main problem was the purity of our samples; principally, freedom from natural mercury contamination. Our procedure is a slight modification of the methods used by Wien and Alvarez⁶ and by Bitter *et al.*¹ Our target was a 0.005-inch gold strip of commercial grade of high purity. It was heated for two hours to 1000°C under vacuum to remove as much natural mercury and other contaminations as possible. After the bombardment was completed, the gold target was cut in smaller pieces and sealed in a quartz boiler. It was then baked at 200°C for two hours under vacuum; this operation removes from the gold target all natural mercury that has adsorbed on its surface during the bombardment stage, and unless it was performed our cells were always seriously contaminated.

Once the gold target is cleaned, it is melted with an induction heater or an oxygen torch; the melting releases the mercury, which is then caught on a clean contorted piece of gold inserted in the pumping lead. With some care it is easy to "catch" 100% of the

radioactive mercury on the clean gold without losing any. Finally, the mercury can be transferred from the "catcher" to the cell by moderate heating (200–300°C).

To discriminate between the various radioactive isotopes present, we used a 256-channel γ -spectrum analyzer, and it was easy to identify the Au¹⁹⁸, Hg¹⁹⁷, and Hg^{197*} peaks. The energy of the beam in the target was from 14.6 to 7.2 Mev, and the following relative yields were obtained:

Hg^{197*} \approx 17%; Hg¹⁹⁷ \approx 54%; Hg¹⁹⁶ \approx 17%; Hg¹⁹⁸ \approx 12%.

Another problem that arose in connection with the samples was the behavior of the quartz cells. Under the intense radiation from the sample, approximately 15 millicuries (mC), the quartz acquired a purple color; this was caused by F centers, since under moderate

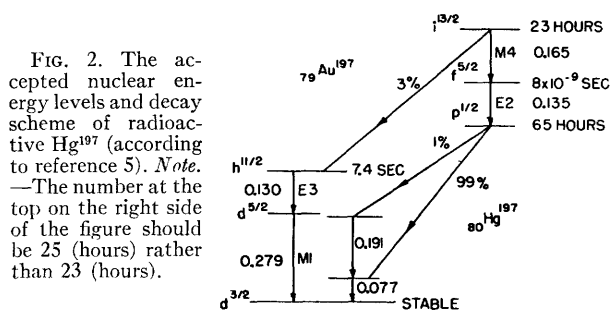


FIG. 2. The accepted nuclear energy levels and decay scheme of radioactive Hg¹⁹⁷ (according to reference 5). *Note.*—The number at the top on the right side of the figure should be 25 (hours) rather than 23 (hours).

⁶ J. Wien and L. W. Alvarez, Phys. Rev. 58, 1005 (1940).

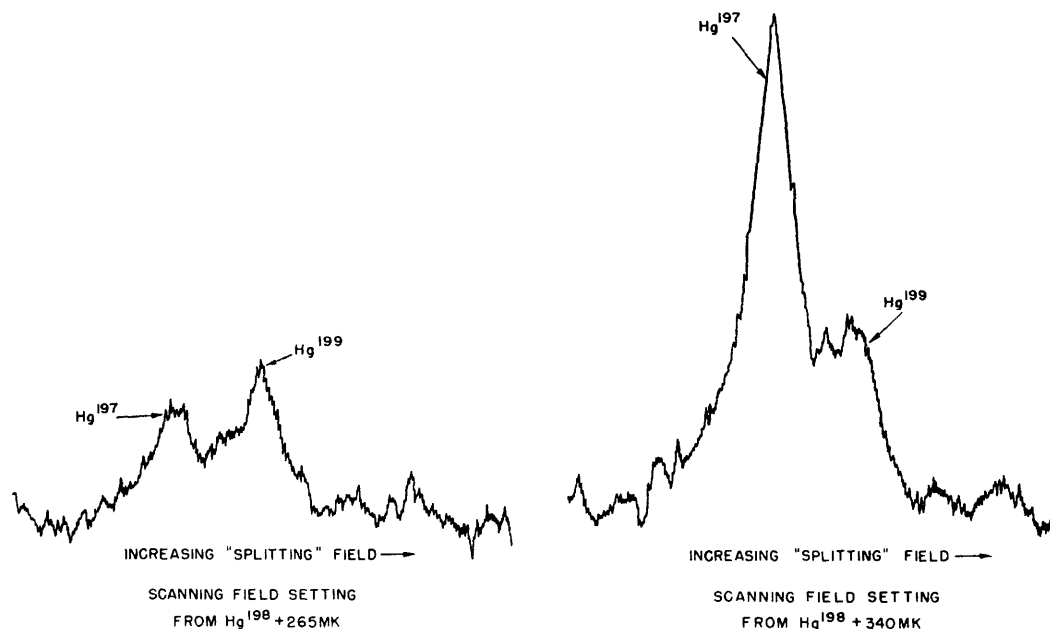


FIG. 3. The $F = \frac{3}{2}$, $m = +\frac{1}{2} \rightarrow m = +\frac{3}{2}$ resonance of Hg^{197} and Hg^{199} . Scanning field set at +265 mK and at +340 mK.

heating the original transparency was restored. What was worse was the release in the cell of large amounts of foreign gas (mainly hydrogen) which completely quenched the resonance radiation. Thus we had to prepare cells with small amounts of radioactive material; the strongest sample to be successfully used was only 1 mC; this amounts to approximately 1.2×10^{13} atoms, and to a corresponding density of 4×10^{12} atoms/cm³ in our cell. Thus we could not reach the optimum density for our geometrical configuration (as determined with Hg^{198}), which was 10^{13} atoms/cm³. (More details on the preparation of the samples are given in the author's Ph.D. thesis.⁷)

EXPERIMENTAL ARRANGEMENTS AND RESULTS

The experimental arrangement is essentially the same as the one described in reference 3, the only difference being that the microwaves were modulated at 30 cps. The detection was achieved by means of a narrow-band phase-sensitive (lock-in) detector. The detector was of the "diamodulator" type,⁸ and was capable of effective bandwidths of the order of 0.01 cps, while 0.1 cps was commonly used. A commercial ferrite isolator, and a variable water-glycol attenuator⁹ were used in the microwave line with satisfactory results.

The microwave frequency was 3053.2 Mc/sec, and the two resonances were observed at "splitting field" values of 2,081.5 and 2,384.7 gauss, respectively. The results for these resonances are summarized in Table I,

where we also give the value of the splitting field for the corresponding Hg^{199} resonances.

Even though the splitting field for Hg^{197} and Hg^{199} is very close, it is well outside the experimental error of the proton resonance measurement used for the field determination. As a matter of fact, in a cell contaminated with natural mercury it was possible to observe the Hg^{197} and Hg^{199} resonances simultaneously (Fig. 3).

The resonance signal obtained for the $m = +\frac{1}{2} \rightarrow m = +\frac{3}{2}$ transition from an uncontaminated Hg^{197} cell is shown in Fig. 4. The signals obtained at various settings of the scanning field have been superimposed to show the construction of a scanning curve. The center of the scanning curve gives the position of the initial m -sub-level (in this case, the $m = +\frac{1}{2}$).

To obtain the value of the nuclear dipole interaction constant, we apply the formula¹⁰

$$A = (\Delta y^2 - \Delta y H) / (H - \frac{3}{2} \Delta y),$$

where $H = g_J \mu_0 B$ in cm⁻¹, B being the splitting field, and $\Delta y = f_{\text{Hg}}/c$, where f_{Hg} is the microwave frequency.

TABLE I. Summary of experimental results.^a

Transition	Isotope	Micro-wave frequency (Mc/sec)	Splitting field (gauss)	Scanning of field (mK)	Zero-field position of $F = 3/2$ level (mK)
$m = -1/2 \rightarrow m = -3/2$	Hg^{197}	3053.2	2081.5	302.4- ₄ ⁺⁷	349.8
$m = -1/2 \rightarrow m = -3/2$	Hg^{199}	3053.2	2076.7		
$m = +1/2 \rightarrow m = +3/2$	Hg^{197}	3053.2	2384.7	408.7- ₇ ⁺⁴	345.0
$m = +1/2 \rightarrow m = +3/2$	Hg^{199}	3053.2	2393.0		

^a 1 mK (millikayser) = 10^{-3} cm⁻¹.

⁷ A. C. Melissinos, Ph.D. thesis, Massachusetts Institute of Technology, 1958 (unpublished).

⁸ Chance, Hughes, MacNichol, Sayre, and Williams, *Waveforms*, Radiation Laboratory Series (McGraw-Hill Book Company, Inc., New York, 1949), Vol. 19.

⁹ D. Alpert, *Rev. Sci. Instr.* **40**, 779 (1949).

¹⁰ A. C. Melissinos, Quarterly Progress Report, Research Laboratory of Electronics, Massachusetts Institute of Technology (January 15, 1957), p. 28.

This gives the following results: from the $m = -\frac{1}{2} \rightarrow m = -\frac{3}{2}$ resonance, $A = 513.0 \pm 2$ mK [where 1 mK (millikayser) $\equiv 10^{-8}$ cm⁻¹]; from the $m = +\frac{1}{2} \rightarrow m = +\frac{3}{2}$ resonance, $A = 513.8 \pm 1$ mK. We accept $A_{197} = 513.5 \pm 1$ mK, and since $A_{199} = 491.5 \pm 0.5$ mK,^{3,11}

$$\mu_{197} = \frac{I_{197} \times A_{197}}{I_{199} \times A_{199}} \mu_{199} = 0.527 \pm 0.001 \text{ nm},$$

where we adopted $\mu_{199} = 0.5043$ nm from reference 11.

The magneto-optic scanning data (see Table I) provide the location of the $F = \frac{3}{2}$ level with respect to Hg¹⁹⁸ (+347.5 mK); thus, we calculate the isotope shift of the center of gravity of Hg¹⁹⁷ from Hg¹⁹⁸ to be $+91 \pm 5$ mK. Further, because our samples contained Hg¹⁹⁶ in a high relative concentration (17%), even-isotope resonance was easily established in Hg¹⁹⁶, and the isotope shift of Hg¹⁹⁶ was measured as $+137 \pm 4$ mK.

DISCUSSION OF THE RESULTS

From the results reported, it is seen how powerful the "double resonance" method is in the case in which only extremely minute samples are available. Indeed, the fact that a paramagnetic resonance signal was obtained with as few as 3×10^{12} atoms is gratifying. An attempt to obtain resonances from the isomeric atom Hg^{197*} has not yet been successful, mainly because of the high nuclear spin ($I = 13/2$), but the hfs has been investigated spectroscopically.¹²

Table II gives the isotope shifts in the 2537 Å line, obtained by the "double resonance" and "scanning"

TABLE II. Isotope shifts in the 2537 Å line of natural mercury.

Isotope	Isotope shift (mK)	Isotope pair	Relative shift in pair	
			200-202 shift	200-202 shift (from reference 13)
196	+137			
197	+ 91	196-198	0.75	0.74
198	0	197-199	0.58	...
199	- 16	198-200	0.85	0.90
200	-156	199-201	1.09	...
201	-216	200-202	1.00	1.00
202	-339			
		202-204	0.98	0.99
204	-519			

¹¹ J. Blaise and H. Chantrel, J. phys. radium **18**, 193 (1957).

¹² A. C. Melissinos and S. P. Davis, following paper [Phys. Rev. **115**, 130 (1959)].

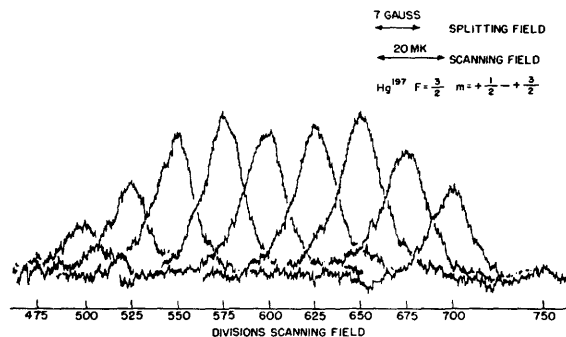


FIG. 4. Superposition of double resonance signals from the $F = \frac{3}{2}$, $m = +\frac{1}{2} \rightarrow m = +\frac{3}{2}$ transition, to show the construction of the scanning curve for the $F = \frac{3}{2}$, $m = +\frac{1}{2}$ sublevel.

combination, of a total of eight mercury isotopes. In column 4 we give the ratio of the even-even and odd-odd isotope shift differences with respect to the 200-202 interval; in column 5 we give the same ratios according to a recent compilation by Brix and Kopfermann,¹³ mainly from other mercury lines. It is seen that even after the even-odd staggering is neglected, large isotope shift anomalies still prevail, which might yield useful information about the electric charge distribution of these nuclei.

Finally, we want to mention that with the present apparatus we made measurements of the lifetime of the 3P_1 state of Hg¹⁹⁸, using the method described in reference 2. We performed our measurements at vapor pressures corresponding to 0°, 13°, 25°, 37°, and 61°C and found consistently at all temperatures $T_e = (1.2 \pm 0.2) \times 10^{-7}$ sec. This is in disagreement with the findings of Guichon *et al.*,¹⁴ but we attribute this lack of "pressure narrowing" to the small dimensions of our cell ($1 \times 1 \times 1$ cm).

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¹³ P. Brix and H. Kopfermann, Revs. Modern Phys. **30**, 517 (1958).

¹⁴ Guichon, Blamont, and Brossel, J. phys. radium **18**, 99 (1957).