IV. NUCLEAR MAGNETIC RESONANCE AND HYPERFINE STRUCTURE

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A. SPINS, MOMENTS, AND ISOTOPE SHIFTS IN NEUTRON-DEFICIENT MERCURY ISOTOPES

A thesis with this title has been submitted by W. J. Tomlinson III to the Department of Physics, Massachusetts Institute of Technology, July 18, 1963, in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

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B. SPIN AND NUCLEAR MOMENTS OF Hg^{203}

Optical spectroscopic measurements were made of the splittings in a magnetic field of one Hg^{203} hyperfine-structure component in the 2537 Å ($6s\,6p\ ^3P_1$-$6s^2\ ^1S_0$) Hg line in absorption. From these measurements a spin assignment was made, and a set of mutually dependent values of the magnetic dipole and electric quadrupole hfs interaction constants A and B was determined.[1] Additional data needed to determine the values of A and B was obtained from measurements of Hg^{203} hyperfine structure in 2537 Å, 4047 Å, ($6s7s\ ^3S_1$-$^3P_0$) and 4358 Å ($^3S_1$-$^2P_1$) Hg emission lines. The high-resolution spectra were obtained with the use of a 10-in. plane diffraction grating (M.I.T. No. 97) mounted in a 36-ft focal-length mirror monochromator.

In earlier hfs investigations of other radioactive mercury isotopes, transmutation reactions were used to produce Hg isotopes of sufficiently high isotopic purity and quantity ($\approx 10^{13}$ atoms) for spectroscopic work with electrodeless lamps. Only pile-produced Hg^{203} (ratio of Hg^{203} to natural mercury $\approx 3 \times 10^{-4}$) was available for this experiment, since adequate quantities were not obtainable by transmutation reactions. Generally it is not possible to measure weak hfs lines in the presence of strong ones if the ratio of intensities is in excess of 100:1 (because the stronger line produces a diffraction pattern that has an intensity approximately 1/100 of the line in the region of extent of the hyperfine structure); hence this Hg^{203} could not be used for a study of emission spectra. Pile-produced Hg^{203}, however, was adequate for measurements in the absorption spectrum in which this difficulty is not incurred. Some emission-spectrum measurements were made possible through the use of Hg^{203} enriched electromagnetically at the Argonne National Laboratory, in which the ratios of Hg^{203} to the other mercury isotopes present ranged from $\approx 1/10$ to ≈ 1. Sufficient mercury was obtained to fill successfully two electrodeless lamps with approximately 10^{14} atoms. The presence of other Hg isotopes in

these lamps made the measurement of all the Hg^{203} hfs components impossible, because of overlap with stable Hg components.

For the absorption measurements, a pressure-broadened Hg^{198} lamp was used as the source for the continuum background. The absorption cell, 10 cm long and 14 mm in diameter, was placed in a magnetic field and heated to ≈100°C. This was found to be the optimum temperature for producing sufficient Hg^{203} absorption without producing an excessive width of the natural Hg absorption. The cell contained approximately 5×10^{14} Hg^{203} atoms.

A spectrum of the 2537 Å line was obtained in emission. It revealed only one new hfs component, removed approximately 0.5 cm^{-1} from the stable Hg hyperfine-structure spectrum. In view of the high intensity of this component, if the hfs of Hg^{203} is resolved, any other components must be overlapped by the natural mercury hfs. Because of this fortuitous position of the isolated Hg^{203} component with respect to the stable mercury, it was apparent that the magnetic-field splitting of the observed line could be studied in an absorption spectrum experiment by using unenriched Hg^{203}. The absorption line was observed, first, at zero field, and then magnetic-field splitting measurements were made at 17 values of the field between 2250 and 10,200 gauss. Also, five observations were made in the low magnetic-field region, 0-2250 gauss, where the components were not

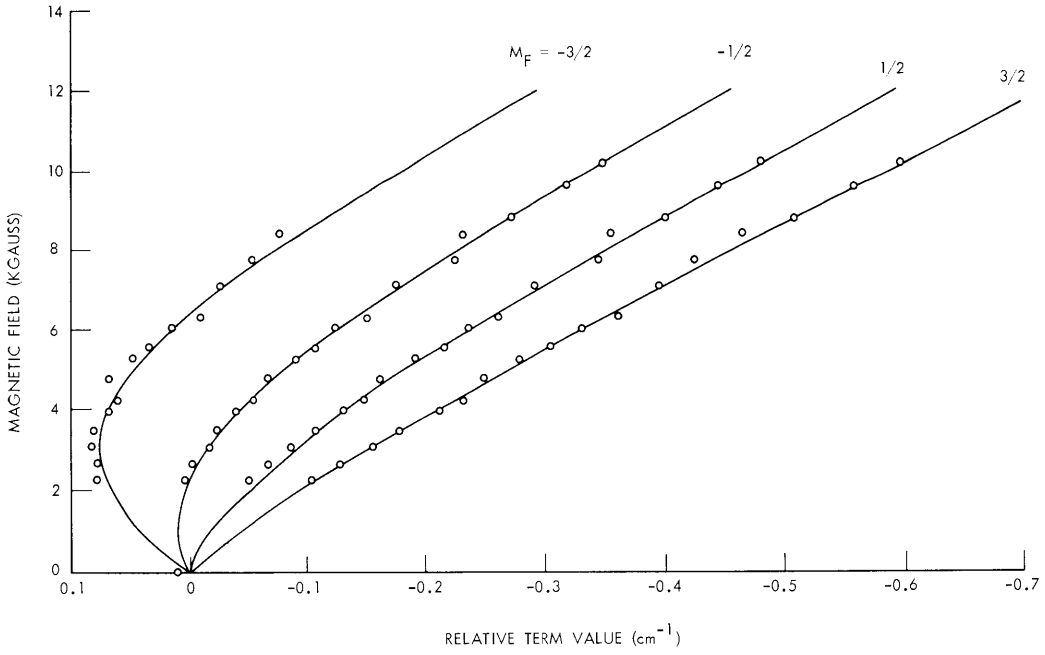

Fig. IV-1. Magnetic-field dependence of the $^3P_1$, F = 3/2 level of Hg^{203} drawn for A = 0.165 cm^{-1}, B = -0.010 cm^{-1}. The experimental points are indicated. Error limits for individual points are approximately ±0.005 cm^{-1}.

resolved. The splitting as a function of the magnetic field is shown in Fig. IV-1.

From the observed splittings it is concluded that with only four M_F components, the observed Hg^{203} hfs component originates from an F = 3/2 level, where $\vec{F} = \vec{I} + \vec{J}$, with $\vec{I}$ the nuclear spin, and $\vec{J}$ the electronic angular momentum (J=1). The spin assignments I = 1/2 and I = 3/2 are definitely excluded, since the qualitative behavior of their F = 3/2 levels as a function of the magnetic field is completely different. For example, for I = 1/2, F = 3/2 (and in fact for any F = I + 1 level) there are two straight lines with slope $\mu_o g_J$ (with $\mu_o \equiv$ Bohr magneton, and $g_J \equiv$ electronic g-factor) for outer components, and for I = 3/2, F = 3/2, the highest energy component would always have the same sign of slope and would not display the change of sign observed at ~3000 gauss (Fig. IV-1).

The possibility that more components exist in the magnetic-field pattern than were detected, because of (a) an intensity that is too low, or (b) overlapping of several components, was investigated and eliminated on the basis of the observable intensities, the linewidths of the components, and the positions of the other hfs components. We concluded therefore that the only consistent spin assignment is I = 5/2.

Our results for the magnetic dipole and electric quadrupole hfs interaction constants A and B are:

$$A = 0.165 \pm 0.004 \text{ cm}^{-1}$$

$$B = -0.010 \pm 0.015 \text{ cm}^{-1}.$$

The magnetic dipole moment obtained from the known ratio of μ (Hg^{201}) to A (Hg^{201}) is

$$\mu = 0.833 \pm 0.020 \text{ nm}.$$

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References

1. The computations were carried out on the IBM 7090 computer at the Computation Center, M.I.T., by using the program "Hyperfine 3" supplied by D. H. Zurlinden, Atomic Research on Radioactive Atoms — Hyperfine 3, Lawrence Radiation Laboratory, University of California, Berkeley, August 1, 1960.

C. MAGNETIC SCANNING OF MERCURY 193

Two unsuccessful attempts have been made to optically pump Hg^{193} (6 hour) and determine its nuclear magnetic moment. A number of scanning curves, however, have been obtained (see Fig. IV-2) which show the hyperfine structure in the $^3P_1$ level of several mercury isotopes. These curves were obtained by varying the magnetic field in which an Hg^{202} lamp is placed and comparing the 2537 Å light scattered at 90° by radioactive mercury vapor in a quartz cell with the 2537 Å output of the lamp. The scanning curves can be compared with the spectroscopic measurements of Tomlinson.[1] The vertical lines below the curve indicate the location and approximate intensity expected according to Tomlinson. A comparison between the scanning peaks and the spectral

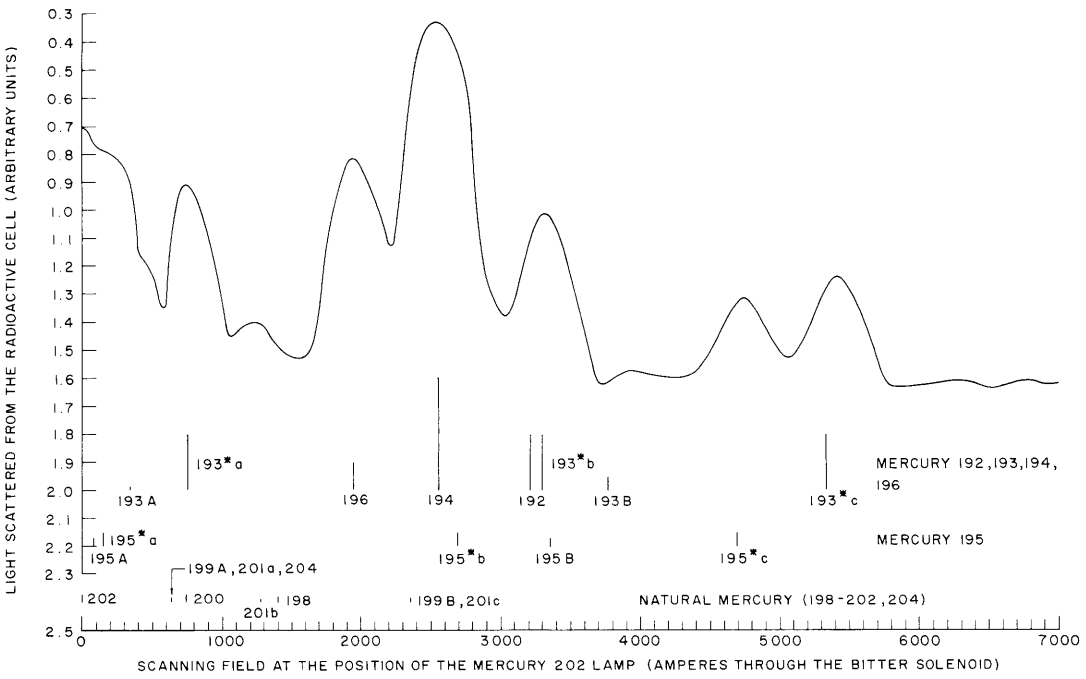

Fig. IV-2. Magnetic-scanning curve of Hg^{193}.

lines is also made in tabular form below. The agreement is good for all of the mercury isotopes present except Hg^{193}.

Component	Scanning Peak		Spectral Line
193^{*a}	740 amp	-289 ± 10 mK	-294 mK
201b	1260 amp	-170 ± 10 mK	-160 ± 4 mK
198			-137 ± 4 mK
196	1950 amp	0 mK	0 mK
194	2560 amp	148 ± 10 mK	147 ± 7 mK
192	3320 amp	332 ± 10 mK	304 ± 20 mK
193^{*b}			326 mK
193B	3970 amp	490 ± 20 mK	442 ± 10 mK
195^{*c}	4740 amp	677 ± 10 mK	665 ± 5 mK
193^{*c}	5420 amp	842 ± 10 mK	821 ± 6 mK
Interval			
196-202		473 ± 10 mK	474 ± 4 mK
195^{*c}-193^{*c}		165 ± 14 mK	156 ± 2 mK
193B-195^{*c}		187 ± 22 mK	223 ± 15 mK

As far as the structure of the scanning curve is concerned, the zero-field peak, for the most part, is due to the large change in lamp intensity when a field is applied. The small wiggles above 6000 amps were not reproducible and should be neglected. The peaks at 1260 amps and 3970 amps, however, were observed in every one of the five scanning curves taken and, therefore, are presumably real. The 1260-amp peak is probably due to a small trace of natural mercury contaminant, while the 3970-amp peak is probably the $Hg^{193}B$ (that is, F = 3/2) component.

Except for Hg^{193}, the agreement between the scanning and spectroscopic results is good. Only the last peak (193^{*c}) falls outside of the combined error limits. This result may be due to a small error in the ampere-to-gauss calibration of the magnet, or to a small departure from linearity at high fields.

The significance of these magnetic-scanning results is, first, that they confirm the 2537 Å results of Tomlinson for Hg^{192}, Hg^{193*}, Hg^{194}, Hg^{195*}, and Hg^{196}, and, second, they suggest that the $Hg^{193}B$ component is displaced farther from the Hg^{196} component than the spectroscopic results indicate. If the scanning data are correct, then both the isotope shift and the staggering parameter γ of Hg^{193} would fall into line with the results of the other mercury isotopes. The isotope shift of Hg^{193} relative to Hg^{196} would be 213 ± 26 mK, instead of 165 ± 20 mK, and thus place it within the error limits of the Hg^{193*} isotope shift (231 ± 6 mK). The staggering parameter γ would be 1.16 ± 0.39 instead of 1.77 ± 0.43. A γ of 1.0 is within the error limits of this value, and thus the Tomlinson-Stroke explanation[2] of the odd-even staggering effect in isotope shift would hold for Hg^{193}, as well as for the other odd mercury isotopes. Notice that it would no longer be necessary to consider a possible nuclear spin of 3/2 for Hg^{193} to reduce γ to 1.0.

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References

1. W. J. Tomlinson III, Spins, Moments, and Isotope Shifts in Neutron Deficient Mercury Isotopes, Ph.D. Thesis, Department of Physics, M.I.T., 1963.

2. W. J. Tomlinson III and H. H. Stroke, Phys. Rev. Letters 8, 436 (1962).