IV. NUCLEAR MAGNETIC RESONANCE AND HYPERFINE STRUCTURE

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A. PARAMAGNETIC RESONANCE IN ATOMS ALIGNED BY ELECTRON IMPACT

Observation of resonances between the magnetic sublevels of the metastable $6^{3}P_{2}$ state in the even isotopes of mercury was previously reported by us (1). The apparatus used in that experiment has been rebuilt for the purpose of scanning the hyperfine-structure levels of the $6^{3}P_{2}$ configuration of Hg¹⁹⁹ and Hg²⁰¹ in order to determine the polarization of these levels and to observe transitions ($\Delta F = 0$; $\Delta m_{F} = \pm 1$) in these levels. Polarization and resonances have now been observed in the even isotopes, Hg¹⁹⁸ and Hg²⁰²; the F = 5/2 and F = 3/2 levels of Hg¹⁹⁹; and in the F = 7/2 and F = 5/2 levels of Hg²⁰¹.

We are using essentially the same apparatus that is described in reference 1 except that the performance of the various components has been greatly improved, and the Hg^{198} lamp has been placed in a magnetic field for the purpose of scanning the hyperfine-structure levels of the odd isotopes.



Fig. IV-1. Recorder traces of polarization of λ 5461 A absorption by (a) $6^{3}P_{2}$ state of Hg¹⁹⁸ atoms, and (b) F = 3/2 level of $6^{3}P_{2}$ state of Hg¹⁹⁹ atoms, plotted against magnetic field. Figure IV-1 shows typical resonance curves at 50 mc in the F = 3/2 ($g_F = 1.8$) level of Hg¹⁹⁹ and in Hg¹⁹⁸ ($g_J = 1.48$). The curves were observed with a radiofrequency power of 0.5 watt and a vapor pressure of mercury of approximately 2 microns. The halfwidths of these curves were approximately 0.2 gauss and were found to depend appreciably on the rf power that was used.

By using an rf power of 0.03 watt (the smallest power with which resonances could be observed) and a vapor pressure of approximately 2 microns, resonance curves with a halfwidth of 0.1 gauss were obtained in Hg^{202} . This corresponds to a lifetime of 1.5×10^{-6} sec of the atoms in the $6^{3}P_{2}$ state in our discharge tube.

At present, we are studying the effects of the discharge-tube current and mercury vapor pressure on the lifetime of the $6^{3}P_{2}$ state. We are also determining the g_{F} values of the F = 5/2 and F = 3/2 levels in Hg¹⁹⁹, and the F = 7/2 and F = 5/2 levels in Hg²⁰¹.

B. B. Aubrey, L. C. Bradley III

References

1. B. B. Aubrey and L. C. Bradley III, Quarterly Progress Report, Reseach Laboratory of Electronics, M.I.T., Oct. 15, 1957, p. 22.

B. HYPERFINE STRUCTURE OF RADIOACTIVE Hg¹⁹⁷ AND Hg^{197^{*}}

1. Preparation of Radioactive Mercury Samples

Radioactive Hg¹⁹⁷ and Hg^{197*} have been prepared by this group for several years. The technique is now fully developed, and fair amounts of pure radioactive mercury (with practically no contamination) are being produced.

A cyclotron bombardment for 12 hours at 40 μ amp produces approximately 10 millicuries of Hg¹⁹⁷ and approximately 3.5 millicuries of Hg^{197*} (about 10¹⁴ and 10¹³ atoms, respectively). The production of Hg^{197*} has been verified by the gamma-ray spectrum of the prepared samples that is shown in Fig. IV-2. The peaks (A-E) are associated with the following transitions:

A - Escape peak from the 77 kev line

- B = 77 kev gold X-ray from the K-capture and the transition to the ground state of Au¹⁹⁷, which is converted by 0.76 ± .10
- $C=138\ \text{kev}$ from the E2 transition in mercury, converted by $0.68\ \pm\ .05$
- D 195 kev transition in gold, converted by 0.7

E = 279 kev from the cascade in gold, converted by 0.4.

The lack of any peak at 165 kev, corresponding to the M4 transition from Hg^{197^*} , which is alarming at first sight, is justified because this line is known to be completely converted (1, 2).



Fig. IV-2. The gamma spectrum of radioactive Hg^{197} and Hg^{197*} produced by 14-mev deuterons on gold by the Au(d, 2n) Hg reaction.



Fig. IV-3. Infrared spectrometer curves of quartz samples.

The number under each peak gives its integrated intensity (total number of counts in the peak). The assignments given above are also justified by the decay rates. As to the purity of the samples, both spectroscopic and double-resonance data indicate the presence of only very minute amounts of natural mercury.

Another problem that was encountered in the preparation of the samples was the radiation damage to the cell. Whenever strong samples were prepared, a strong discoloration of the quartz appeared. Furthermore, when the cells were sealed at pressures of 10^{-4} mm Hg, the vacuum deteriorated to 1-2 mm within a few hours. Very strong hydrogen lines were detected spectroscopically, and we suspect that hydrogen is knocked out of the quartz by beta and gamma radiation. Naturally, such pressures completely quench the resonance radiation. We hope that this serious difficulty will be overcome by use of quartz of the proper quality. As an example, the infrared spectra of samples of General Electric Company and of Ammersil quartz are shown in Fig. IV-3. The General Electric quartz shows a very strong absorption band at 2.65μ because of the presence of OH.

Thanks are due Professor R. C. Lord, Director of the Spectroscopy Laboratory, M.I.T., for the infrared spectra. All of the delicate glass blowing and handling of radioactive material was done by Mr. A. J. Velluto, Research Laboratory of Electronics, M.I.T., whose ability has been an important factor in the successful preparation of our samples.

A. C. Melissinos

References

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- 2. C. H. Braden, L. D. Wyly, and E. T. Patronis, Jr., Phys. Rev. <u>95</u>, 758 (1954).

2. Magnetic Dipole Moment and Isotope Shift of Radioactive ${\rm Hg}^{197}$

Double-resonance signals in this isotope were observed for the first time a year ago and were reported in the Quarterly Progress Report of July 15, 1957, page 41. However, much stronger signals have recently been obtained with the use of a phase-sensitive detector. The results are as follows.

For the magnetic dipole interaction constant of the ${}^{3}P_{1}$ level of Hg¹⁹⁷ from the $m = -1/2 \rightarrow -3/2$ transition, we obtain a = 513.0 ± 2 × 10⁻³ cm⁻¹; from the $m = +1/2 \rightarrow +3/2$ transition, we obtain a = 513.8 ± 2 × 10⁻³ cm⁻¹; and we accept a = 513.5 ± 1 × 10⁻³ cm⁻¹.

For the zero-field location of the F = 3/2 level of Hg¹⁹⁷ from the m = -1/2 scanning curve, we obtain $349.8^{+7}_{-4} \times 10^{-3}$ cm⁻¹; from the m = +1/2 scanning curve, we obtain



Fig. IV-4. Superposition of double-resonance signals to show the construction of the F = 3/2, m_F = +1/2 scanning curve.



Fig. IV-5. Scanning curve for the F = 3/2, $m_F = -1/2$ sublevel.



Fig. IV-6. Scanning curve for the F = 3/2, $m_F = +1/2$ sublevel.

 $345.2_{-7}^{+4} \times 10^{-3} \text{ cm}^{-1}$; and we accept $347.5 \pm 5 \times 10^{-3} \text{ cm}^{-1}$. Hence the isotope shift of the center of gravity of Hg¹⁹⁷ from Hg¹⁹⁸ is +90.8 ± 5 × 10⁻³ cm⁻¹.

Figure IV-4 is a superposition of double-resonance signals obtained for various settings of the scanning field. It shows both the quality of the double-resonance signals and the scanning curve that is the envelope of these signals.

Figures IV-5 and IV-6 give the scanning curve for the m = -1/2 and m = +1/2 sublevels of Hg¹⁹⁷. These measurements were performed with 0.3 millicurie of radioactive mercury; that is, with only 10¹² atoms (approximate) of Hg¹⁹⁷.

These measurements conclude the present work on the ${}^{3}P_{1}$ level of Hg¹⁹⁷. However, a direct microwave resonance experiment at 23,085 mc between the F = 1/2 and F = 3/2 levels now seems possible if sufficient power can be obtained.

A. C. Melissinos

3. Spectroscopic Study of Radioactive Hg^{197} and Hg^{197*}

In a program designed for gaining further insight into the static properties of nuclei, we have intensified the study of the shift in the spectral lines of atoms which results from a change in the nuclear charge distribution of a single atom (1). The possibility of performing such experiments is offered by studying the spectra of certain atoms in the nuclear ground state and in a reasonably long-lived isomeric level.

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Theoretical work on this subject was initiated by Weiner (2). His treatment is based essentially on the single-particle model and predicts large values (compared with accuracies that can be obtained spectroscopically) for odd proton nuclei and small values for odd neutrons. For the latter he ascribes any measurable effects to the electronneutron interaction. We expect (3), however, that the main contribution to the isomeric shift with odd neutrons will come from the nucleon-core coupling. In fact, we can study this interaction in a simpler situation than in the isotope shift, in which different numbers of nucleons are involved. Isotopes of xenon, as well as of mercury, are being studied.

A discharge tube containing approximately 3 millicuries of 65-hour radioactive Hg^{197} was prepared (approximately 4 × 10¹³ atoms). From one-third to one-fifth of the atoms were in the 25-hour isomeric state (Hg^{197*} , I = 13/2), as evidenced by the gamma-ray spectrum (see Section IV-B 1).

The tube was filled with argon at relatively high pressure (approximately 10 mm Hg) and excited with a 200-mc oscillator. It performed well, but after each 15-minute period of operation it had to be flamed out because the mercury had cleaned up (was driven into the walls). As the mercury was gradually driven into the walls molecular bands appeared with increasing intensity. No evidence of natural-mercury contamination could be found.

The tube was studied in emission with a Fabry-Pérot interferometer crossed with a fast spectrograph. Two etalons were used (spacers, 2.993 mm and 4.33 mm), and the following lines were analyzed:

λ4047	$\left(65\ 6p\ ^{3}P_{0}-65\ 7s\ ^{3}S_{1}\right)$
λ4078	$\left(\begin{array}{cccccccccc} 6 \text{ s 6p }^{3} \text{P}_{1} \text{ - 6s 7s }^{1} \text{S}_{0} \end{array}\right)$
λ 4358	$\left(65\ 6p\ ^{3}P_{1}-65\ 7s\ ^{3}S_{1}\right)$

Exposures from 20 to 30 minutes were used.

Echelle pictures were also taken, and the following lines were analyzed:

$$\lambda 2537$$
 $\left(6s^{2} {}^{1}S_{0} - 6s 6p {}^{3}P_{1}\right)$
 $\lambda 4047$
 $\lambda 4358$

The exposures varied in duration from 30 minutes to 1 hour.

In all lines, the Hg¹⁹⁷ components appeared strong and well resolved. The hyperfine splittings of Hg¹⁹⁷ in the ${}^{3}P_{1}$ (770 mk) and in the ${}^{3}S_{1}$ (1135 mk) are exactly as calculated from the paramagnetic resonance in the ${}^{3}P_{1}$ level (see Section IV-B 2).

The Hg¹⁹⁶ component was present in all of the lines at the correct position. This







Fig. IV-7. (a) Echelle spectrogram of $\lambda 2537 \text{ A}$ of radioactive Hg^{197} , Hg^{197*} , 24 hours after bombardment. (b) Fabry-Pérot spectrogram of $\lambda 4078 \text{ A}$ of radioactive Hg^{197} , Hg^{197*} , 24 hours after bombardment.





SPACER 2.993MM



Fig. IV-8. Echelle and Fabry-Pérot spectrograms of λ 4047 A of radioactive Hg¹⁹⁷, Hg^{197*}, 24 hours after bombardment.





was expected because the bombarding energy is above the 11-mev threshold of the Au(d, 3n)Hg reaction, and Hg¹⁹⁶ is produced with Hg¹⁹⁷. Furthermore, several weak components appeared in all of the lines. They do not belong to natural mercury, to Hg¹⁹⁶, or to Hg¹⁹⁷; they are attributed to the isomeric state of Hg^{197*}. All of these components vanished after three days when exposures of as long as 1 hour were taken, except for the +817-mk component in the 4047 line, which persisted but was extremely weak.

The complete analysis of the two etalon spectrograms, in conjunction with the echelle pictures, uncovered the structure of the four lines, as given in Figs. IV-7a, IV-7b, IV-8, and IV-9.

The energy-level assignments of Hg¹⁹⁷ are also given in these figures. However, an attempt to make energy-level assignments for Hg^{197*} did not seem feasible. For example, the intensities of the two components in $\lambda 2537$ A indicated a positive moment for Hg^{197*}; but the two unassigned components in $\lambda 4047$ indicated – by their position – a negative moment, unless an extremely large isomeric shift in the ${}^{3}S_{1}$ level exists. Moreover, the energy-level scheme for the ${}^{3}P_{1}$ level cannot be constructed consistently from $\lambda 4078$ and $\lambda 2537$, both of which lead to it.

Therefore we have decided to postpone further analysis of the energy-level scheme of Hg^{197*} (and of the isomeric isotope shift) until more data are collected on the structure of the lines.

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References

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C. INTENSITY OF ULTRAVIOLET RADIATION OF A MICROWAVE-EXCITED MERCURY PLASMA IN A MAGNETIC FIELD

In the study of the hyperfine structure of the 2537 A mercury line, electrodeless discharges in magnetic fields are used. It has been known that the intensity of the emitted ultraviolet radiation varies with the magnetic field; but the variation is not monotonic, and consistently, the curve for an increasing field differs from the curve for a decreasing field.

In this experiment (our data are shown in Fig. IV-10), the intensity was measured every 500 gauss up to 10,000 gauss. "Steps" appear to occur at 2000 gauss and 6000 gauss, while at 8000 gauss saturation seems to have been reached. The continuous curve



Fig. IV-10. Intensity of $\lambda 2537A$ from Hg¹⁹⁸ plasma excited at 3000 mc in a magnetic field.



Fig. IV-11. Calibration of 4-inch Bitter solenoid.

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gives the results for increasing fields; the dotted curve, for decreasing fields. Note that the sharp increase at 6000 gauss has a corresponding decrease on the dotted curve at 5000 gauss. The curve is drawn for both directions of the magnetic field in order to show the reproducibility of the results.

The source was an electrodeless discharge of Hg^{198} with 1 mm of argon, excited by 10-cm microwaves. Although the power delivered to the magnetron was constant, the power delivered to the load was not monitored. We did, however, measure the temperature of the vycor envelope of the source in order to gain some information about the vapor pressure of the mercury. We found that the temperature of the external side of the envelope varied in exact proportion to the intensity variations that are shown in Fig. IV-10.

A. C. Melissinos

D. VARIATIONS IN THE CALIBRATION OF HIGH-CURRENT SOLENOIDS

Since high-current "Bitter" solenoids (1) are now in extensive use, it should be reported that their calibration — field versus current — is not constant. As we expected, it depends upon the clogging of the cooling ducts in the solenoid because temperaturedistribution variations change the current distribution in the copper plates.

In Fig. IV-11 calibration curves for the 4-inch solenoid used by our group are given for November 1953, March 1956, February 1957, and, after the cooling system had been cleaned, for May 1958. It is also worth mentioning that the most homogeneous spot of the solenoid was shifted 1 inch between the last two calibrations.

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References

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