Optical Studies of Radioactive Mercury*

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(Received July 6, 1954)

A technique has been developed for studying the hyperfine structure of the resonance radiation of mercury. A single Zeeman component of the 2536 A line of Hg^{198} is used as a variable-frequency monochromatic light source, and the field strength applied to the arc lamp to produce resonance absorption in a resonance lamp is a measure of the resonance frequency. Natural mercury, and radioactive mercury produced by bombarding gold with 15-Mev deuterons were investigated in this way and also (by means of an echelle spectrograph) in the emission spectrum of an electrodeless discharge. Several observed lines have not been conclusively identified.

The magnetic moment of Hg^{197} is 4 percent greater than that of Hg^{199}. The isotope shift of isotopes with even mass number decreases with decreasing mass number. This effect may be due in part to the increasing quadrupole moment with neutron deficiency in these isotopes.

THE work reported below is part of a general program of exploration of atomic energy levels in search of information about nuclear structure that is revealed by the interaction of the nucleus with the atomic electrons. For the past year and a half we have been developing methods and techniques applicable to mercury because of the ease with which the famous ultraviolet resonance line at 2537 A and other lines originating on various metastable levels may be excited, because hyperfine structure and isotope shift are readily measurable, and because a whole series of radioactive isotopes having half-lives of the order of hours are available for investigation. We had hoped, by this time, to have obtained more data, and more accurate data, than is reported in this paper. It has now become quite clear that to reach many of our objectives we shall have to construct new apparatus, which will incidentally facilitate the observations in which we have been engaged. It was therefore decided to report our progress and to summarize our experiences and results at this stage.

As a first project, we set ourselves the task of investigating the hyperfine structure of the resonance radiation of radioactive isotopes of mercury. Resonance radiation was chosen because, in principle at least, it could be investigated with only enough atoms to fill a small cell at some very low pressure, whereas other spectral lines would require an emission spectrum produced in an electrical discharge that might drive the mercury into the cell walls, and thus require more atoms to produce observable effects for many hours of observation. Moreover, a "magnetic scanning" technique (described below), ideally suited to the investigation of the hyperfine structure of very small quantities of mercury, was being developed in our laboratory.[1] We therefore set out, first of all, to establish our requirements by studying the hyperfine structure of the resonance radiation of natural mercury.

HYPERFINE STRUCTURE OF RESONANCE RADIATION OF NATURAL MERCURY

A summary of experimental data on hyperfine structure and isotope shift, compiled by Brix and Kopfermann, is contained in a section of the 1952 edition of the Landolt-Börnstein tables in the volume dealing with nuclear data. This is an excellent guide to the abundant literature on the subject. The material dealing with the resonance radiation of mercury may be divided into two main categories. The first relates to emission spectra analyzed by conventional optical equipment. The outstanding reference to work of this kind dealing with the resonance line of mercury is by Schüler and Keyston.[2] The second category is concerned with absorption spectra. Mrozowski[3] showed that if resonance radiation from a natural mercury lamp is passed through a mercury absorption filter in a strong magnetic field, all but one of the hyperfine structure components are removed from the beam. A variety of investigators, particularly Buhl,[4] used this single line to investigate the hyperfine structure of mercury in a very ingenious way. It was allowed to fall on a resonance lamp containing natural mercury in a magnetic field. As the field was varied, the frequencies of various Zeeman components of different hyperfine structure lines successively reached the frequency of the single incident line. Whenever this condition was obtained, resonance radiation was removed from the incident beam and was reradiated in all directions. From a knowledge of the fields for which this occurred, the zero field pattern could be reconstructed by using the available auxiliary data about the approximate locations and g factors of the various lines and levels.

* This work was supported in part by the Signal Corps; the Office of Scientific Research, Air Research, and Development Command; and the Office of Naval Research; in part by a U. S. Atomic Energy Commission Contract.

† Work performed while a National Science Foundation fellow.

[1] Bitter, Plotkin, Richter, Teviotdale, and Young, Phys. Rev. **91**, 421 (1953).

[2] H. Schüler and J. E. Keyston, Z. Physik **72**, 423 (1931).

[3] S. Mrozowski, Acad. Polonaise Sci. et Lettres Bull. No. 9–10A, pp. 464, 1930, Bull. No. 6A, pp. 489, 1931.

[4] O. Buhl, Z. Physik **109**, 180 (1938); and **110**, 395 (1938).

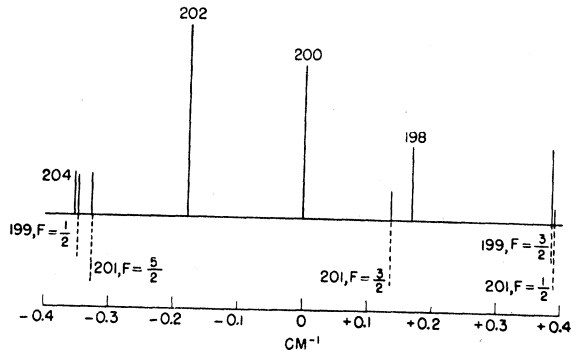

FIG. 1. The hyperfine structure of $\lambda = 2537$ A of mercury.

Brix and Kopfermann conclude from a review of all available experimental data that the best present estimate for the structure of the 2537 A line of mercury is that shown in Fig. 1. We shall return to a comparison of these results with our own in the last section of this paper, and content ourselves at this point with a qualitative review of the main features.

The levels involved in the emission of the resonance line are the $^3P_1$ excited state and the $^1S_0$ ground state. The ground state has no electronic magnetic moment and is therefore always single, regardless of the nuclear angular momentum. Since mercury nuclei contain an even number of protons, nuclei with an even mass number will contain an even number of neutrons and may be expected to have $I=0$. The resonance lines from these even isotopes will therefore be single lines. The positions of these lines for mass numbers 198, 200, 202, and 204 are shown in Fig. 1. Hg^{196} is present to only 0.15 percent and had not been observed in natural mercury. The height of the vertical lines for the even-even isotopes is proportional to the abundance of the isotope. Hg^{199} has a spin $I=1/2$, and the excited state splits into two, with $F=1/2$ and $3/2$. The resulting two lines are shown, one near each end of the pattern, in Fig. 1. Hg^{201} has a nuclear spin of $3/2$, and the hyperfine structure pattern contains the three lines shown. The typical observed structure consists of five lines of comparable intensity four of which are approximately equally spaced, the fifth on the short wavelength side being somewhat more separated from its neighbor. The structure of the unresolved lines is inferred from data on other lines, and from Zeeman effect investigations.

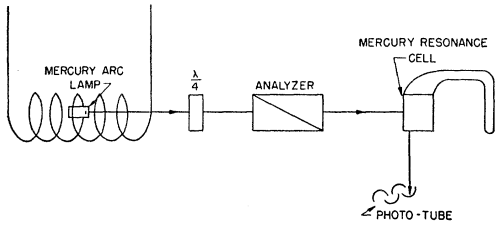

FIG. 2. Diagram of the apparatus used for magnetic scanning.

DESCRIPTION OF APPARATUS USED IN MAGNETIC SCANNING EXPERIMENTS

The main features of our apparatus are shown schematically in Fig. 2. A mercury arc lamp containing only the 198 isotope is placed in a magnetic field, and a beam is formed in the direction of the field. The Zeeman effect for an even isotope consists of a classical triplet with only the two outer circularly polarized components radiated in the direction of the beam. By means of a quarter-wave plate and a suitably oriented analyzing prism, either one of the two frequencies present may be suppressed. The apparatus provides a single line of variable frequency, the frequency being determined by the orientation of the prism and the magnitude and sense of the magnetic field applied to the arc, or scanning field, as we shall call it. This light is allowed to fall on a resonance lamp containing the sample to be investigated, and the emission of resonance radiation is detected with a galvanometer and photomultiplier. The frequency at which resonance is observed may then be calculated from the strength of the scanning field and the known g factor of the even isotopes of mercury.

The arc consisted of a vycor tube, approximately 5 mm in diameter, containing argon at a pressure of a few millimeters and the Hg^{198}, and was excited by means of a 200-Mc/sec oscillator to give an electrodeless discharge. Filled tubes approximately 6 inches long were supplied by Baird Associates. They were cut with a torch into two or three pieces in order to fit into the magnet used; it was found that all the pieces could be used as light sources. Since some of these pieces must have contained mostly vapor, and little if any condensed mercury, this was an indication that an electrodeless discharge of the kind used requires very little excess mercury. Abnormal line shapes were occasionally encountered. These were presumably due to the self-reversal of the resonance line. They were completely eliminated by water-cooling the arc.

The scanning magnet was an air-core solenoid having an inside diameter of 4.5 inches and being capable of producing fields up to 50 kilogauss.[5] Since the field in this magnet is not exactly proportional to the magnet current, and depends on the temperature distribution in the magnet, it was monitored by a Laboratory for Electronics nuclear resonance magnetometer. Although this magnet was far from ideal for our purpose, it was used for this preliminary work. The field fluctuated irregularly, sometimes by as much as 50 gauss. It was sufficiently uniform to see resonances on an oscilloscope only when the probe was within an inch of the center. For the most precise measurements the probe was kept at the center, and the arc lamp was placed $1\frac{1}{8}$ inches above the probe. In this way the relative magnitudes of the fields used could be accurately determined. The absolute magnitude at the lamp was estimated to be

[5] F. Bitter, Rev. Sci. Instr. **10**, 373 (1939).

2.5 percent lower than the field at the magnetometer probe.

The light from the lamp was focused into a parallel beam by means of a quartz lens, and then passed through a crystalline quartz quarter-wave plate, and a Glazebrook prism cemented with glycerine. A small brass rod, not shown in Fig. 2, could be inserted into this beam to scatter light into an auxiliary photo tube to monitor the lamp intensity.

The plane-polarized monochromatic beam illuminated a quartz cell placed between the poles of a small iron core magnet capable of producing fields up to 10 kilogauss. This made possible the investigation of the Zeeman effect of the resonance radiation. The resonance lamp cells were made for us by the Neider Fuzed Quartz Company. They were in the shape of small cubes having edge lengths ranging from 0.5 inch to 1 inch. One face was drawn out into a tail and could be immersed in a low-temperature bath. Although these lamps usually behaved satisfactorily, anomalous line shapes attributable to causes that we have not investigated in detail were occasionally observed. They may have been associated with the presence of condensed mercury at the lamp faces, since the procedure of flaming the faces of the lamp and insuring that condensed mercury remains in the tail seems to have cured these difficulties.

In some of the first lamps that we filled with radioactive mercury, the resonance radiation disappeared even though they had appeared to be quite satisfactory immediately after filling. Flaming sometimes made the resonance radiation disappear completely. This effect was unquestionably the result of some kind of chemical cleanup associated with impurities retained in the liquid air traps used to hold the mercury during the filling process while the cells were being evacuated. It was found that the mercury could be selectively held on clean gold foil at room temperature during the pumping process. Then, after the cell was sealed off, the mercury could be removed from the gold by gentle heating. The cell was, of course, carefully baked out before filling. These precautions sufficed to remove the troubles with chemical cleanup of the resonance cells.

The resonance radiation from these cells was either focused on an IP28 photomultiplier by means of another quartz lens, or was "conducted" to it by means of a glass tube, 0.5 inch in diameter, silvered on the inside. The photomultiplier current could be either measured directly with a galvanometer, or balanced by the current from the auxiliary photomultiplier, mentioned before, in an optical bridge.[6] For quick but less accurate observations the potential across a shunt carrying the scanning magnet current was applied to the horizontal plates of an oscilloscope with a dc amplifier and the vertical plates to a resistor carrying the photomultiplier current. This could be done, since the magnet power supply was not grounded. The field could then be varied from +10 kilogauss to −10 kilogauss in a half-minute or so, and the oscilloscope trace photographed with a Land camera, yielding pictures of the hyperfine structure of the resonance radiation like that shown in Fig. 3. This was an extremely useful technique in establishing interesting conditions for more detailed study with the galvanometer.

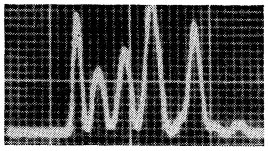

FIG. 3. Oscilloscope trace of the hyperfine structure of the resonance radiation of natural mercury.

In some of our experiments it was found helpful to eliminate the light caused by fluorescence of the quartz cell by placing a Corning 9863 filter in front of the photomultiplier.

TYPICAL RESULTS

Typical results obtained with the apparatus described above are shown in Fig. 4. The upper curve is for natural mercury, and was obtained with the resonance cell at room temperature. The five main peaks expected

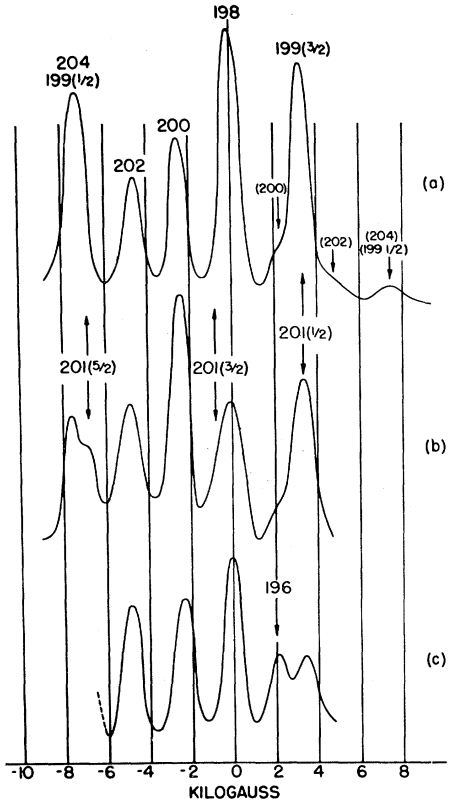

FIG. 4. The hyperfine structure of the resonance radiation from three samples of mercury having different isotopic compositions.

[6] O. Oldenburg and H. P. Broida, J. Opt. Soc. Am. **40**, 381 (1950).

are clearly shown. In addition, there are three small partly resolved peaks on the right side of the figure. These are also visible in the oscilloscope trace in Fig. 3. They are attributable to the fact that the light transmitted by the quarter-wave plate is not completely plane-polarized, and the Glazebrook prism, consequently, does not quite completely eliminate one of the two radiated frequencies. They are reflections of the peaks to the left of the central 198 peak.

The maximum of the 198 peak is slightly to the left of the zero field line. This is presumably caused by the presence of the small unresolved 201(3/2) peak.

The positions of these peaks will be discussed in the last section of this paper, their magnitude in the next section, in which we take up the separation and identification of overlapping peaks. The width of these lines at half maximum is about 1 kilogauss or, using a conversion factor derived further on, 0.063 cm^{-1}. Their Doppler width at room temperature would be 0.035 cm^{-1}.

The middle curve in Fig. 4 was obtained with a sample of mercury enriched in the 201 isotope, furnished by the Atomic Energy Commission. Its composition was 37.3 percent even isotopes, 4.76 percent Hg^{199}, and 57 percent Hg^{201}. The corresponding percentages for natural mercury are 69.8 percent, 17.0 percent, and 13.2 percent. The 5/2 peak of 201 on the left is quite apparent, and the 3/2 peak is beginning to make an appreciable bulge on the side of the 198 peak.

The lower curve in Fig. 4 was obtained from the residues of a radioactive resonance lamp some months after it had been prepared. A new peak appears on the right side of this diagram. It is caused, as we shall show, by stable Hg^{196}, which was produced along with radioactive Hg^{197} in the cyclotron.

SEPARATION AND IDENTIFICATION OF OVERLAPPING LINES

It was hoped that we might identify and locate all the lines present by producing a Zeeman effect in the resonance cell and following each line as a function of applied field as it crossed other lines in the pattern. This still is a possibility, but the work is extremely time-consuming and tedious except for the isotopes of even mass number. The central π-component of these lines is undisplaced by a magnetic field, whereas the π-components of the lines resulting from the odd isotopes are not independent of field strength. By using incident radiation polarized with its electric vector parallel to the auxiliary Zeeman field, it is possible to be certain that a line comes from an isotope of even mass number, and second, to remove any overlying lines by a suitable choice of field strength. This was done in identifying and measuring the position of the line due to the even isotopes.

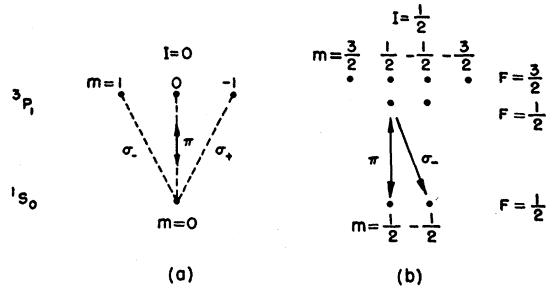

FIG. 5. Transition diagram for the even isotopes and Hg^{199}.

The lines from isotopes of odd mass number are more difficult to handle. If the lines to be measured are well separated from their neighbors, it is, of course, possible to split them, count the number of components, and measure their g factor. But when the lines are faint and are covered by other lines, it was found difficult to carry out such a procedure with certainty. At one point we hoped to simplify the patterns to be studied by excluding some lines on the basis of their polarization. Thus, for example, if we excite one of the even isotopes with light polarized parallel to the splitting field, this will necessarily put the atom in an excited state with $m=0$, and the only possible radiative transition back to the ground state is the correspond-transition shown in Fig. 5(a). If we exclude the π component in the resonance radiation either by means of an analyzing prism, or by observing in the direction of the field, we should expect to find the resonance radiation suppressed, particularly if we use sufficiently low vapor pressures so that depolarizing collisions are avoided. For the isotopes of odd mass number, on the other hand, exciting with π radiation should not eliminate σ_- resonance radiation, since there are two possible transitions to the ground state, one π and one σ_- as shown in Fig. 5(b). The expected effect was in fact not realized to a sufficient extent to make this scheme practically useful for selectively suppressing the lines caused by the even isotopes.

An aspect of our results not yet brought out is that the relative intensities of the various peaks are a function of the vapor pressure determined by the temperature of the tail of the lamp. Each peak has its maximum height at some temperature which is different for the various peaks. As pointed out by Mrozowski,[7] geometrical factors may be very important in determining the temperature at which the maximum is obtained. We have made no investigation of this point.

A particularly marked instance of effects resulting from this maximum in the intensity of the resonance radiation was observed in a cell containing 90 percent Hg^{204}. This sample was also supplied to us by the U. S. Atomic Energy Commission. At room temperature there was no 204 peak at all, but all of the other peaks, caused by minor amounts of the other isotopes present, showed up clearly. When the tail of the lamp was cooled to $-30°C$, the 204 peak alone was present, and the

[7] S. Mrozowski, Phys. Rev. **93**, 641 (1954).

others were too faint to observe. Unfortunately, this selective suppression of peaks is effective only when concentration differences approach 100 percent.

The problem of isolating and identifying overlapping peaks resulting from isotopes of odd mass number is now being worked on, and we hope to report on practical usable methods in the not too distant future.

PREPARATION OF RADIOACTIVE MERCURY

The mercury isotopes were prepared by bombarding a 4 mil thick gold foil with 15-Mev deuterons in the M.I.T. cyclotron. The thickness of the gold foil was such that the energy of the deuterons after traversing the foil would be reduced to near the threshold of the reaction $Au^{197}(d,2n)Hg^{197}$. This energy-loss requirement on the gold thickness was set to reduce Hg^{198} contamination produced in the reaction

$$Au^{197}(d,p)Au^{198}\rightarrow\beta^{-}+Hg^{198}$$

for the cross section for this reaction is well below its maximum value at the threshold of the $(d,2n)$ reaction, and decreases with increasing energy.

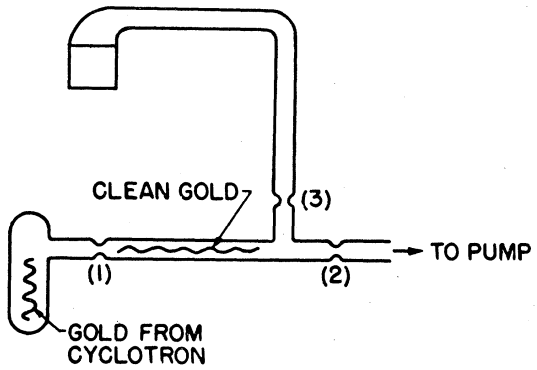

FIG. 6. The distillation apparatus used to separate radioactive mercury from gold.

Before irradiation, mercury surface contamination was removed from the gold by heating in a hydrogen atmosphere for one and a half hours at a temperature of 900°C. As soon as the gold had cooled, it was placed in the cyclotron.

The gold was bombarded with deuterons at a beam current of 40 μa for a period of 12 hours. An additional period of four hours was allowed for the very intense, short-lived activity in the aluminum target-backing to die out. The gold was then cut from the target and placed in the quartz vacuum system illustrated in Fig. 6. After the vacuum system had been pumped down, the target gold was heated to just above its melting point with an induction heater, and the mercury distilled off. The mercury was then caught on the surface of another piece of clean gold which had been previously heated to drive out occluded gases and placed in the vacuum system. The system was then sealed off at point (1), allowed to pump down, and then sealed off at point (2). The resonance cell was then placed in liquid air, and the "catching" gold gently heated to drive the mercury off its surface and into the cell. The cell was then sealed off at point (3), and was ready for use.

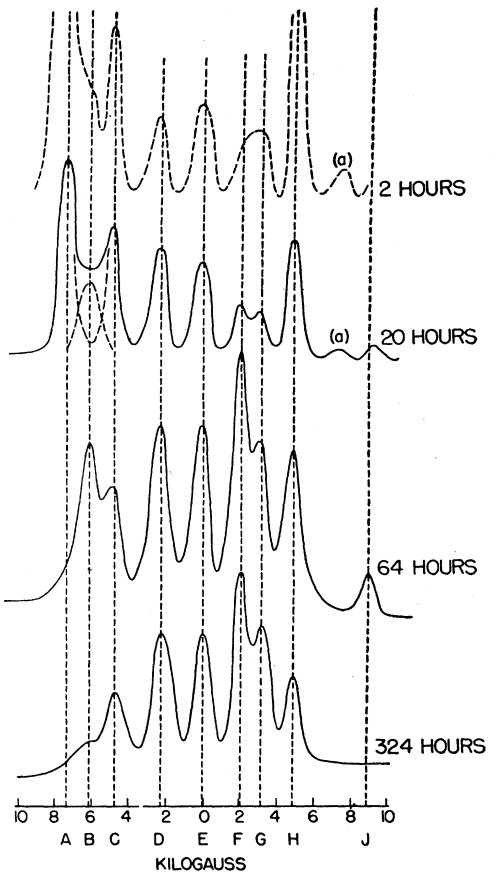

FIG. 7. The hyperfine structure of the resonance radiation of radioactive mercury at various times.

HYPERFINE STRUCTURE AND ZEEMAN EFFECT OF RESONANCE RADIATION OF RADIOACTIVE SAMPLES

The first run with radioactive mercury in February 1953 was successful. A large number of curves were recorded, of which a representative selection is shown in Fig. 7. At least nine resolved lines were observed. About a dozen runs have been undertaken since that time with the object of identifying these various lines. Many of these runs yielded no results at all because of faulty techniques. However, enough work has been done with the apparatus described to indicate that the point of diminishing return has been reached, and that a new approach is required if further progress is to be made without undue expenditure of time. The results of our efforts to identify the observed lines are summarized below.

The procedure used to identify the peaks was to observe the splitting when the resonance lamp was

placed in a magnetic field, and to make observations both with the electric vector of the incident radiation parallel to the field (i.e., with the π component used for illumination) and with the electric vector of the incident radiation normal to the field (i.e., with the σ component used for illumination).

In none of the runs after the first was the A peak as pronounced during the early hours as for this first run. Also, in none of the subsequent runs did it disappear completely. We conclude that there is, here, a peak attributable to a radioactive isotope falling at the same place as the peaks resulting from the 204 isotope and the $F=1/2$ peak of the 199 isotope.

The B peak is, apparently, also caused by a radioactive isotope, since it comes at a point for which no peak is observed in natural mercury and disappears completely in the spectrum of the residues. It was observed to split into two components in a magnetic field with either π or σ illumination. This behavior is consistent only with the assignment of $F=1/2$. Furthermore, the g factor was measured by observing the magnitude of the separation of the two peaks in several known fields. The measured g factor was consistent only with the assignment of $F=1/2$. We conclude that the B peak must be attributable to an isotope with a nuclear angular momentum $I=3/2$, or $1/2$, since only these values can lead to the required $F=1/2$ in the $^3P_1$ state in which $J=1$.

The peaks C, D, and E come at the places to be expected for the 202, 200, and 198 isotopes. The changing relative magnitudes are not enough in themselves to justify assigning any of these peaks to a radioactive isotope. The G peak comes at the point where the 199 $F=3/2$ peak is to be expected. If the G peak is from 199, it is difficult to understand the absence of the corresponding $F=1/2$ peak at A in the 324-hour curve of Fig. 7. If it is not from the 199 isotope, then it is hard to understand the presence of the 202 and 200 peaks, which could not possibly have been made by bombarding Au^{197} with deuterons and must therefore be caused by contamination. We must be satisfied for the present with the negative statement that there is no conclusive evidence for the presence of radioactive peaks at C, D, E, and G, although it is perfectly possible that these peaks may completely or partly overlap peaks due to radioactive isotopes.

Peak F is surely attributable, at least in part, to the stable isotope 196. It is unchanged in strong fields when π illumination is used and is split into two components when σ illumination is used. It is found in the spectra of the residual vapor after radioactivity has decayed, as shown in Fig. 4(c). It must therefore come from a stable isotope with $I=0$, and this can only be the isotope 196.

The peak at H is caused by a radioactive isotope. It is split into two components by a magnetic field when π illumination is used, but when σ illumination is used two additional components appear. This is consistent with the assignment of $F=3/2$, as is the measured value of the g factor. We conclude that this line must be due to a nucleus with $I=5/2$, $3/2$, or $1/2$.

The splitting of the peak at J was too complex to make an assignment. The number of components was apparently greater than two or four, which should have been easily recognizable for this well-separated line.

Although photographic records were made of the Zeeman splittings produced in a wide range of fields, the confusion of overlapping lines discouraged detailed analysis, and no further conclusions were drawn from the available data.

The location of all of the lines observed is given in Table I in the next section. Although most of the observations were recorded simply as galvanometer deflections for a given magnet current, a series of measurements, particularly for the even isotopes, was made in which the deflection was observed as a function of the proton resonance frequency near the intensity maxima. The optical frequency difference Δf in cm^{-1} is then given in terms of the observed proton resonance frequency f_p measured in cycles per second by the relation

$$\Delta f=\frac{g_{\mathrm{Hg}}}{g_p}\frac{M_p}{m}\frac{f_p}{c}.$$

The values of the constants used in applying this relation were 1836 for the ratio of the mass of the proton to that of the electron, 5.585 for the gyromagnetic ratio of the proton,[8] 3.00×10^{10} cm/sec for the velocity of light, and 1.484 for the g factor of the $^3P_1$ state of the even isotopes of mercury.[9] Our results are probably correct to within approximately ±3 percent.

TABLE I. Summary of results. The numbers listed are the displacements of the various lines observed from the Hg^{200} line in cm^{-1}. The numbers given are estimated to be reliable to ±3 percent.

Line	Position according to Brix and Kopfermann	Present investigation
A		−0.343
204	−0.354	−0.343
199(1/2)	−0.350	−0.343
201(5/2)	−0.329	−0.294
197(1/2)		−0.254
202	−0.179	−0.171
201 c.g.	−0.054	
200	0	0
201(3/2)	+0.136	
199 c.g.	0.143	0.136
198	0.168	0.157
197 c.g.		0.245
196		0.293
199(3/2)	0.389	0.376
201(1/2)	0.390	
197(3/2)		0.495
J		0.788

[8] Sommer, Thomas, and Hipple, Phys. Rev. **82**, 697 (1951).
[9] J. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952).

SPECTROGRAPHIC STUDY OF LINE 4047 OF RADIOACTIVE SAMPLES

In connection with the work on radioactive mercury 197, it was thought worth while to examine other levels than those connected with the 2537 A line in order to confirm the values of nuclear spin and magnetic moment as determined from magneto-optic measurements on this line. Since the emission spectrum of mercury was easily obtainable in an electrodeless discharge, and a spectrograph of fairly high resolution was available, this method of attack was tried. The line at 4047 A was chosen because it was due to the relatively simple transition $^3P_0-{}^3S_1$ giving one line for the even isotopes, two lines ($F=1/2$ and $3/2$) for 199, and three lines ($F=1/2$, $3/2$, $5/2$) for 201. The hfs of the line, as given by Schüler and Keyston.[2]

The source was a sealed quartz tube 3 mm by 30 mm, containing the radioactive mercury and argon gas at approximately 7 mm Hg pressure. As determined from the counting rate, there were initially about 2×10^{15} atoms, or 0.5 μg, of radioactive mercury present. Excitation was provided by a 10-cm wavelength magnetron. The tube was placed inside the center conductor at the open end of a coaxial pipe and was cooled with air.

The spectrograph used was a 200 groove/inch Bausch and Lomb echelle, in a Wadsworth mounting.[10] At 4047 A, it has a theoretical resolving power of 330 000 and a plate factor of 0.4 A/mm. All exposures were taken on Eastman type IV-0 spectroscopic plates. Times varied from one to 30 minutes.

Spectrograms were taken beginning about 12 hours after cyclotron bombardment of the gold foil had stopped. For comparison purposes, the spectra of mercury 198 and ordinary mercury were placed on opposite sides of the spectrum of the radioactive sample, overlapping slightly. When first excited, the tube showed the mercury spectrum as well as C_2 and CN bands, as observed in a visual spectroscope. The intensity of the mercury lines increased and that of the impurities decreased as the temperature became higher. The discharge did not fill the tube, unlike all of the previous ones containing ordinary mercury. One attempt was made to apply a magnetic field, but it was unsuccessful because the field extinguished the discharge.

Two spectrograms of the line at 4047 A are shown in Fig. 8. One was taken within one half-life of mercury 197 after bombardment had stopped, and the other at about six half-lives. The upper line in each case is due to the radioactive sample, and the lower to ordinary mercury. Figure 8(b) clearly shows no appreciable difference in the two lines, indicating that the residue from the radioactive sample is ordinary mercury. Figure 8(a), however, shows two important differences

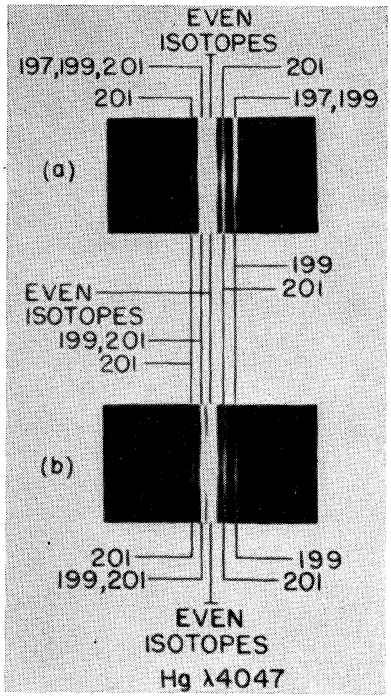

FIG. 8. Photograph of the hyperfine structure of the resonance radiation of two samples of mercury having different isotopic compositions.

in the lines. The relative intensities of the hfs components are not the same, and one component is shifted away from the center, toward longer wavelengths. These spectrograms are consistent with the assumption that the radioactive sample contains some ordinary mercury as a contaminant, and that 197 has a spin of 1/2 (the same as 199) and a magnetic moment slightly greater than 199. The hfs components belonging to 201 and the even isotopes coincide in each photograph, as they should. Only one of the components of 197 is more intense than, and appears to be shifted relative to, those of 199, because the other is overexposed and is not resolved from a component of 201.

Since 199 is still present in the radioactive sample, the center of gravity of the components (slightly different if 197 alone were present) is shifted in the direction of smaller over-all splitting. The measured ratio μ_{197}/μ_{199} is therefore smaller than the true ratio. Measurement and comparison of the relative spacings of the hfs components of 197 and 199, on two different plates, give the ratio $\mu_{197}/\mu_{199}=1.033\pm0.016$.

Spectrograms of other strong lines in the mercury emission spectrum were taken at the same time. They were not used for measurement because of the complexity of their hfs components. The 2537 A line was heavily over-exposed on most plates, and in addition did not have narrow lines, probably because of the high temperature at which the tube was operated.

[10] Harrison, Archer, and Camus, J. Opt. Soc. Am. **42**, 706 (1952). Harrison, Davis, and Robertson, J. Opt. Soc. Am. **43**, 853 (1953).

DISCUSSION OF RESULTS

The expected isotopes in the radioactive samples investigated are Hg^{197} with a half-life of 64 hours, and the isomeric excited state Hg^{197*} with a half-life of 23 hours. (The entire series of neutron deficient mercury isotopes has been investigated by Gillon, Gopalakrishnan, de-Shalit, and Mihelich.[11]) The spins of these isotopes are found to have the values 1/2 and 13/2, respectively. These values are in accord with the single-particle shell model.

Two of the lines observed are compatible with a nuclear spin $I=1/2$. They are B and H of Fig. 7. They are slightly displaced toward higher wave numbers from the lines due to the 199 isotope, which also has a spin $I=1/2$. They are in the same order, that is, the $F=3/2$ line is on the high wave number side. There is nothing in the decay of these lines to indicate that they could not be caused by the same isotope. They are in positions compatible with the independent spectroscopic evidence discussed in the previous section. We therefore have made the assignment indicated in Table I, which contains a summary of all our data. The position of the various lines is given in terms of separation from the 200 isotope to facilitate a direct comparison with the results listed by Brix and Kopfermann in the Landolt-Bornstein Tables.

For natural mercury, the agreement between our results and the summary of previous results is within the experimental error except for the 201 $F=5/2$ line. We consider our result the more reliable since we had, for the first time, an enriched isotope in which this line was partly resolved. We did not attempt a recalculation of the magnetic moment or quadrupole moment of the 201 isotope, since the remaining lines were not resolved by the methods used, but we hope to return to this point in a later communication.

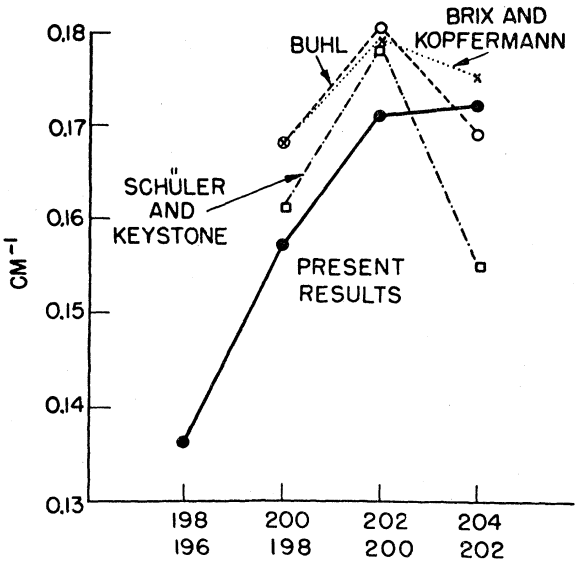

FIG. 9. Isotope shift of the resonance radiation of the even isotopes of mercury. The position of the Hg^{204} peak was obtained with a sample, previously mentioned, markedly enriched in this isotope.

The magnetic moment of the 197 isotope may be computed from the observed hyperfine structure of this and the very similar 199 isotope, and the observed moment[12] of Hg^{199}, which for our purposes may be taken as 0.50 nuclear magnetons. The result is

$$\mu_{Hg^{197}}=0.50\times0.749/0.719=0.50\times1.04$$
$$=0.52 \text{ nuclear magnetons.}$$

The results for the isotope shift of the even isotopes is shown in Fig. 9, where the results of other observers are also plotted. We believe the smooth trend shown by our data to be substantially correct. It is in agreement with the results obtained by various observers using other spectral lines. Of these we quote only Murakawa,[13] who recently published an estimate of the isotope shift for Hg^{196} in a sample in which the natural abundance of 0.15 percent was boosted to 1.46 percent. The observations on the line Hg II λ6150 are tabulated below, together with our own results on isotope shift, using a scale in which the 204–202 displacement is unity.

	204–202	202–200	200–198	198–196
Murakawa	1.0	1.0	0.85	0.60
Present study	1.0	1.0	0.91	0.79

Considering the difficulty of Murakawa's estimate of 0.019 cm^{-1} for the 198–196 displacement in which the 196 line was unresolved but appeared only as a somewhat diffuse edge on the 198 line, the agreement must be considered excellent.

We can hardly hope to discover the meaning of this trend in isotope shift for the theory of nuclear structure without a good deal more evidence. There are undoubtedly departures from the $A^{1/3}$ law for the nuclear radius for various different reasons in different parts of the nuclear "periodic table."

Wilets, Hill, and Ford[14] discuss a variety of reasons for anomalous isotope shifts, of which we shall take up only that due to nuclear electric quadrupole moments, first suggested by Brix and Kopfermann.[15] Mercury, with $Z=80$, lacks two protons of having a closed shell with 82, and the isotope Hg^{204} lacks two neutrons of having a closed shell with 126.

As more and more neutrons are taken out of the closed shell, progressively larger quadrupole moments are to be expected. Exceptionally large quadrupole moments are found in nuclei in which 10 or more nucleons are lacking from closed shells. In this group,

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

[12] W. G. Proctor and F. C. Yu, Phys. Rev. **81**, 20 (1951).

[13] K. Murakawa, Phys. Rev. **93**, 1232 (1954).

[14] Wilets, Hill, and Ford, Phys. Rev. **91**, 1488 (1953).

[15] P. Brix and H. Kopfermann, Physik **126**, 344 (1949). For a further discussion of intrinsic quadrupole moments of nuclei with spin 0 or ½, see also A. Bohr and B. Mottleson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

values of Q/R^2 in the range of 5–10 are observed. Since Hg^{196} lacks 10 neutrons from a closed shell, it is interesting to consider whether the observed trend in the isotope shift might be related to the intrinsic quadrupole moments.

Electronic energy levels are raised due to the finite size of a nucleus by an amount proportional to the mean value of ρr^2 averaged over the nucleus. For a nucleus with quadrupole moment Q and a volume equal to that of a sphere of radius R, the mean value of ρr^2 is proportional to

$$ZR^2[1+(\tfrac{5}{6}Q/ZR^2)^2].$$

The change in an energy level δ resulting from a change in neutron number δN, assuming $R=R_0A^{1/3}$, is

$$\delta=\text{const}\left[\frac{2}{3}\frac{1+(\tfrac{5}{6}Q/ZR^2)^2}{A}+\frac{d(\tfrac{5}{6}Q/ZR^2)^2}{dN}\right]\delta N.$$

If we call the first term δ_R the isotope shift caused by the change in volume, and the second term δ_Q the isotope shift caused by the change in shape, the above expression may be put into the form

$$\frac{\delta_Q}{\delta_R}=\frac{25A}{48Z^2}\delta\left(\frac{Q}{R^2}\right)^2.$$

If we assume the normal volume effect for all the isotope shifts shown in Fig. 9 to be approximately 0.172 cm^{-1}, and the departures from this value to be the decrease δ_Q due to the change in quadrupole moment, we can compute the changes in Q/R^2 from one isotope to another. The effects for 204–202 are too small for us to estimate. However, if we assume a value of Q/R^2 for Hg^{202}, the calculation of Q/R^2 from the data can be carried through for the other isotopes with the results given in Table II. These values appear to be of the correct order of magnitude, and it would therefore seem reasonable to assume that, pending the accumulation of further evidence, the anomalous isotope shift for the even-even mercury nuclei is, at least in part, attributable to the variation of the intrinsic quadrupole moment from one isotope to another.

TABLE II. Assumed and calculated values of Q/R^2.

Assumed value[a] of Q/R^2 Hg^{202}	Calculated value of Q/R^2 from observed isotope shift Hg^{200}	Hg^{198}	Hg^{196}
0.2	0.63	2.41	4.35
0.4	0.72	2.44	4.37
1.0	1.16	2.61	4.46

[a] The experimental value of Q/R^2 for Hg^{201} is 0.6.

ACKNOWLEDGMENTS

We are indebted to Henry Plotkin, who took part in the original experiments leading to the establishment of the magnetic-scanning technique, and who helped us throughout in making observations, and particularly to Alan Teviotdale whose participation in our endeavors was terminated by a fatal accident last summer. His patience and devotion were perhaps the most important factors in overcoming the discouraging difficulties which delayed us for almost a year.

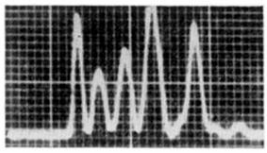

FIG. 3. Oscilloscope trace of the hyperfine structure of the resonance radiation of natural mercury.

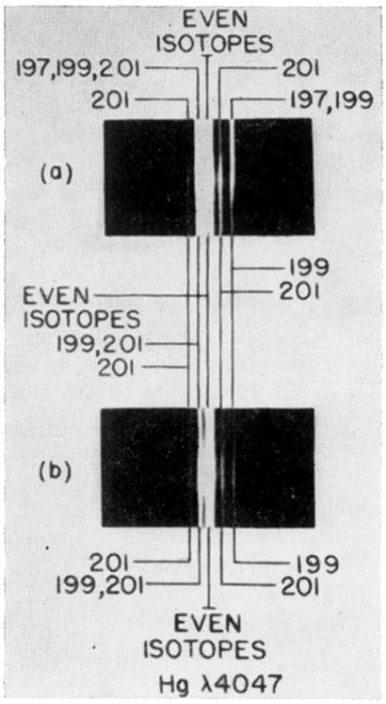

FIG. 8. Photograph of the hyperfine structure of the resonance radiation of two samples of mercury having different isotopic compositions.