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HYPERFINE-STRUCTURE SEPARATIONS, ISOTOPE SHIFTS, AND NUCLEAR MAGNETIC MOMENTS OF THE RADIOACTIVE ISOTOPES Tl^{199}, Tl^{200}, Tl^{201}, Tl^{202}, and Tl^{204}

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Hyperfine-Structure Separations, Isotope Shifts, and Nuclear Magnetic Moments of the Radioactive Isotopes Tl199, Tl200, Tl201, Tl202, and Tl204

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The hyperfine structure and isotope shift of five radioactive isotopes of thallium have been measured spectroscopically. Thallium-199 (7.4 hr) and thallium-200 (27 hr) were produced by alpha bombardment of gold; thallium-201 (3 day), thallium-202 (12 day), and thallium-204 (4 year) were produced by deuteron bombardment of liquid mercury. The isotopes were excited in electrodeless-discharge lamps. Approximately 0.001 μg of radio-thallium was used in the lamps. The hfs splittings and isotope shifts in the $6^2P_{\frac{1}{2}}$, $6^2P_{\frac{3}{2}}$, and $7^2S_{\frac{1}{2}}$ states were obtained by using a 10-in. plane diffraction grating in autocollimation in a 40-ft mirror monochromator. Previously measured values of the nuclear spins of the odd-even isotopes have been verified. Nuclear magnetic moments or upper limits on the moments have been calculated by comparison of the hyperfine splittings of the radioactive isotopes with the known splittings and moments of the stable thallium isotopes. A definite dependence of the relative isotope shifts on neutron number was observed.

I. INTRODUCTION

HYPERFINE-STRUCTURE and isotope-shift studies by optical spectroscopy have been restricted, in general, to nuclei with even-proton even-neutron numbers, and with even-proton odd-neutron or even-neutron odd-proton numbers. It is of interest to study such hfs and isotope shifts in adjacent elements. Kuhn and his co-workers[1–3] have made systematic studies of even-neutron isotopes of medium A nuclei. If we are not to limit our investigations to even-neutron numbers and to elements for which the atomic number Z differs by 2, we are led forcibly to odd-odd nuclei that are radioactive. We want to point out that although the hfs separations in many such nuclei have been obtained by atomic-beam magnetic-resonance experiments, these measurements do not yield the energies of the centers of gravity of the hfs pattern, that is, the isotope shifts. The present experiments are the first instance of isotope-shift studies of unstable odd-odd nuclei.

Since considerable work[4] has already been done on the hyperfine structure of mercury $(Z=80)$, we have investigated a number of isotopes for the adjacent element, thallium $(Z=81)$. Our results therefore permit an isotope-shift comparison between the two sets of isotopes.

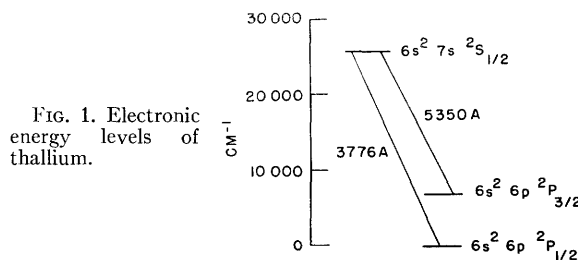The atomic transitions in thallium which we have studied are shown in Fig. 1. The hyperfine structure of

FIG. 1. Electronic energy levels of thallium.

the $^2P_{\frac{3}{2}}$ state is too small to be resolved in these optical experiments.[5] It is therefore simple to interpret the hfs in the lines, since we have to consider only the magnetic dipole interaction (see Fig. 2). The electronic properties of thallium have been discussed in some detail else-

·¹ H. G. Kuhn and S. A. Ramsden, Proc. Roy. Soc. (London) **A237**, 485 (1956).

² W. R. Hindmarsh, H. G. Kuhn, and S. A. Ramsden, Proc. Phys. Soc. (London) **A67**, 478 (1954).

³ H. G. Kuhn and A. G. Warner, Proc. Roy. Soc. (London) **A245**, 330 (1958).

⁴ P. Brix and H. Kopfermann in *Zahlenwerte und Funktionen*, edited by H. H. Landolt and R. Börnstein (Springer Verlag, Berlin, 1952), *I/5* p. 1; F. Bitter, S. P. Davis, and B. Richter, Phys. Rev. **96**, 1531 (1954); J. Blaise and H. Chantrel, J. phys. radium **18**, 193 (1957); A. C. Melissinos and S. P. Davis, Phys. Rev. **115**, 130 (1959); H. R. Hirsch and C. V. Stager, J. Opt. Soc. Am. **50**, 1052 (1960); H. R. Hirsch (J. Opt. Soc. Am., submitted for publication); R. H. Kohler, Phys. Rev. **121**, 1104 (1961); C. P. Brot, Quarterly Progress Report No. 59, Research Laboratory of Electronics, M.I.T., October 15, 1960, p. 54, and J. phys. radium (to be published).
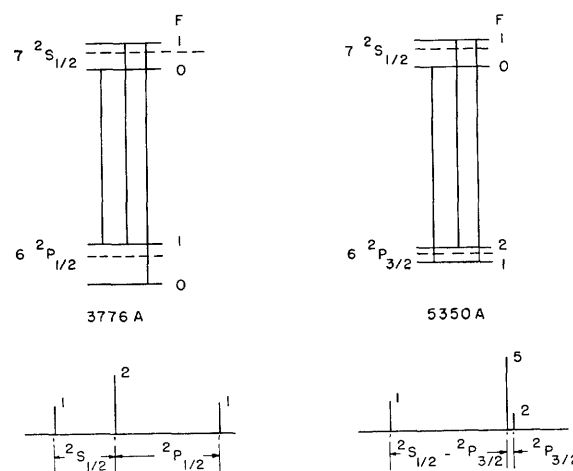
FIG. 2. Hyperfine-structure intervals for a spin $\frac{1}{2}$ Tl isotope. Bottom part of diagram shows the theoretical structure of the lines with relative intensities indicated.

⁵ G. Gould, Phys. Rev. **101**, 1828 (1956).

where[6,7] and we shall proceed directly to the experimental problems.

II. EXPERIMENT

A. General Procedures

1. Sample Analysis

The radioactive isotopes were produced by bombardment of either gold or mercury targets in the M.I.T. cyclotron. Two very small samples were taken from the target, and the remainder was used in the preparation of an electrodeless-discharge lamp. One sample was used to determine the decay curve of the target—this curve being a composite of the half-lives of the various products made by the bombardment. The gamma-ray spectrum of the other sample was taken on a 256-channel analyzer. This spectrum was used as a check on the isotopic species present in the sample by comparing the spectrum with published gamma-ray spectra.
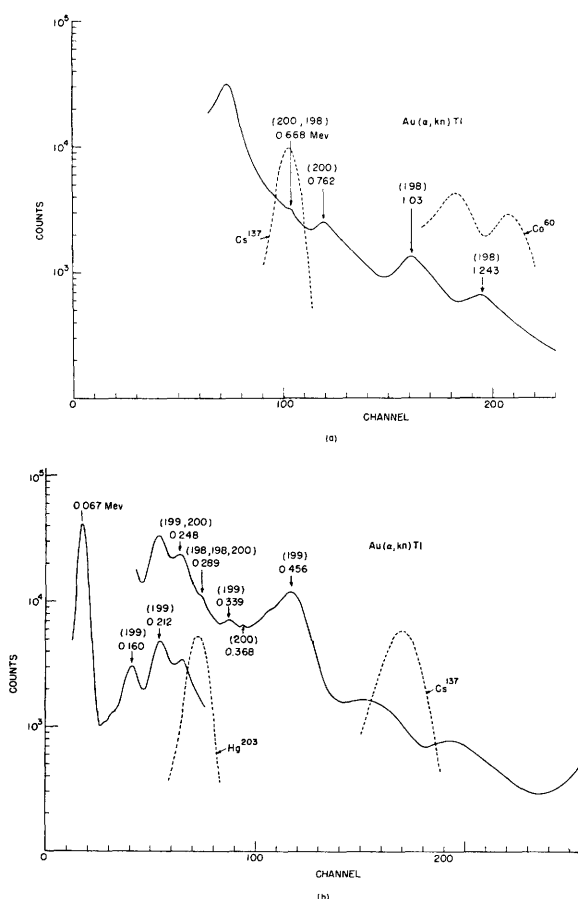

FIG. 3. Typical gamma-ray spectra of the Au (α,kn) Tl bombardments. Hg[203], Co[60], and Cs[137] were used for calibration. (a) High-energy part of the spectrum. (b) Low-energy part of the spectrum.

2. Light Source

The preparation of the lamp will be described in Sec. B. This lamp was used as the light source for the spectrograph to obtain the structure of the thallium 3776- and 5350-A lines. First, a calibration spectrum was placed on the photographic plate. For this purpose, an Osram thallium lamp, cooled by a small blower, was used. Then, the spectrum of the electrodeless lamp that contained radio-thallium was photographed after the Hartmann diaphragm[8] had been shifted. A 50-Mc oscillator, running at approximately 50 w input, was used to excite the lamp. The lamp required external heating to bring out the thallium spectrum. Just enough heat was supplied to show the green line of thallium in either a hand spectroscope or a Hilger D 187 spectrometer. "Just enough heat" means that the lamp was usually run red hot. The Doppler widths at 700°C for the green and the ultraviolet lines are 30 mk and 42 mK $(1 \text{ mK} = 0.001 \text{ cm}^{-1})$, respectively. Of course, there was no self-reversal—even at this temperature—because of the very low density of thallium atoms.

The lamp was supported in a horizontal position between ceramic rods, and a Bunsen burner heated the lamp from underneath. Narrow strips of aluminum foil, to which were clipped the leads from the oscillator, were wrapped around the rods near the lamp. We found very early in this research that metal electrodes in direct contact with a heated quartz lamp will attack the walls and quickly destroy the lamp. A slightly enlarged image of the lamp was focused on the slit of the spectrograph. To increase the irradiance somewhat, a concave mirror was placed behind the lamp. This mirror formed a real image of the lamp at the position of the lamp. Therefore this image was also focused on the slit of the spectrograph. No measurements were made of the actual increase in irradiance, but the image of the lamp on the slit was noticeably brighter with the mirror than without it.

3. Spectrograph

The spectrograph is a mirror monochromator of 40-ft focal length in which a 10-in. M.I.T. plane diffraction grating (No. 97) is used in autocollimation.[9] The grating is blazed to throw approximately 50% of the light into a small range of angles near 63°. It has a groove spacing of approximately 300 grooves/mm. All spectrograms were taken in high orders: usually the 11th for the green line, and the 15th or 16th for the ultraviolet line. The plate factors were approximately 500 mK/mm in these orders.

The spectrograph was modified so that both of the

[6] H. Kopfermann, *Nuclear Moments*, translated by E. E. Schneider (Academic Press, Inc., New York, 1958).

[7] M. F. Crawford and A. L. Schawlow, Phys. Rev. **76**, 1310 (1949).

[8] G. R. Harrison, R. C. Lord, and J. R. Loofbourow, *Practical Spectroscopy* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1948), p. 101.

[9] G. R. Harrison and G. W. Stroke, J. Opt. Soc. Am. **50**, 1153 (1960).

resonance lines could be photographed simultaneously.[10] The grating angle was set to place the ultraviolet line toward one side of the plate holder. Then an extra mirror of 36-ft focal length was placed so as to intercept the green line as it was diffracted from the grating and to focus it on the other side of the plate holder. Since Doppler broadening is proportional to frequency, the slightly shorter focal length of the new mirror did not seriously affect the resolution.

In all cases, Kodak 103a-0 and 103a-F spectroscopic plates were used to photograph the ultraviolet and green lines, respectively. A rotating step sector was placed before the slit to give various exposures. Exposure times were approximately 20 sec for the Osram lamp, and from 2 to 8 hr for the radio-thallium lamps.

B. Target and Source Preparation

1. Thallium-199 (7.4 hr) and Thallium-200 (27 hr)

To produce these isotopes, sheets of chemically cleaned and thoroughly outgassed gold foil ($5 \times \frac{5}{8} \times 0.005$ in.) were bombarded with 30-Mev alpha particles in the external beam of the M.I.T. cyclotron at currents of 5–15 μa for periods of 2–8 hr. The variation in current was due to the unpredictable behavior of the external alpha beam on any given day. Gold consists of a single stable isotope of mass number 197. These external beam runs produced the reactions: $Au^{197}(\alpha,n)Tl^{200}$, $Au^{197}(\alpha,2n)Tl^{199}$, $Au^{197}(\alpha,3n)Tl^{198}$, and $Au^{197}(\alpha,3n)Tl^{198*}$, as verified by a determination of the half-life of the irradiated target and by an analysis of the gamma-ray spectrum (see Figs. 3 and 4). Relatively small quantities of Tl^{198} and its isomer were produced; no photographs

of its spectrum were obtained. Orth et al.[11] indicate that the reactions producing these two isotopes have a maximum at 38 Mev.

After bombardment, the gold was removed from the target holder, cut into quarters and sealed into the vacuum system, together with 1 μg of indium. We found that the addition of 1 μg of In as a carrier increased the useful life of the lamp appreciably (see Appendix). The cell containing the gold and the associated glassware of the vacuum system were baked at 200°C for approximately 2 hr to remove mercury vapor and any other volatile impurities. Liquid nitrogen was then placed on the traps. The gold was melted by using a 5-kw induction heater. Then the radio-thallium, which had evaporated from the gold, was driven into a section of 3-mm quartz tubing by heating the cell containing the gold target with a torch to a bright red heat. The thallium was trapped by means of wet asbestos tape wrapped around the quartz tubing. The cell was sealed off at the joint between the cell and the tubing, and argon was admitted at a pressure of 1–2 mm Hg. An rf discharge was then run on the system with the use of a 50-Mc oscillator for cleaning purposes, as described in the Appendix. With clean argon the pressure was finally adjusted to 1 mm Hg, and the lamp was sealed and pulled off the system.

This was the first, and also the simplest, handling procedure employed, but not all of the desired results were so obtained. The cause of the difficulty was believed to be an insufficient number of atoms in the lamp (see Appendix). Longer bombardments in the cyclotron would have been very difficult because of the general characteristics of the external alpha beam. The
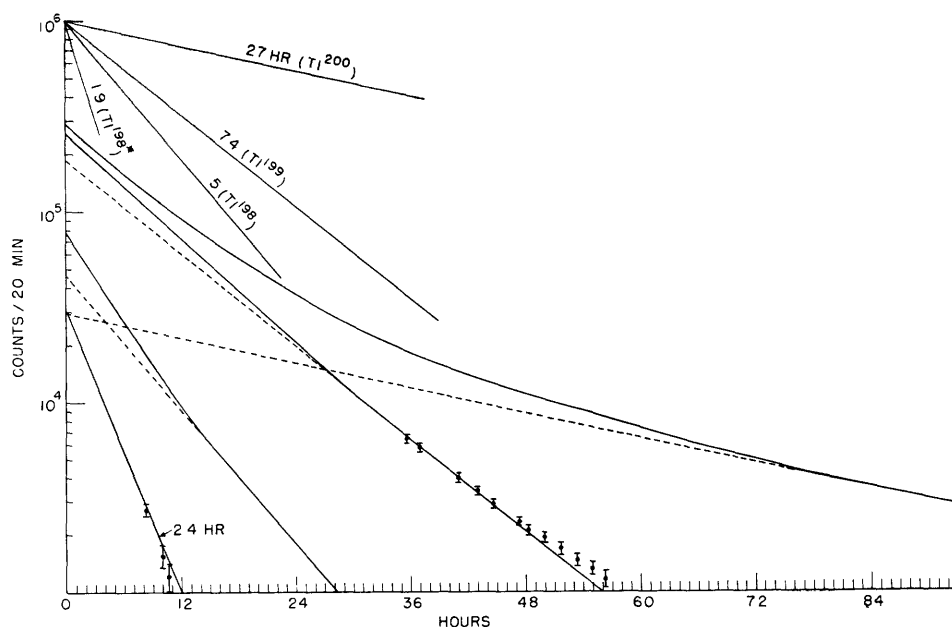
FIG. 4. Half-life analysis of gold target after alpha bombardment. The dashed lines represent the contribution of the isotopes shown in the upper left portion of the figure. The three lower curves are obtained by successive subtraction of the longest-lived component (dashed straight lines) from the original experimental curve.

[10] H. H. Stroke and K. K. Y. Li, J. Opt. Soc. Am. 51, 678 (1961).
[11] D. A. Orth, L. Marquez, W. J. Heiman, and D. H. Templeton, Phys. Rev. 75, 1100 (1949),

GOLD TARGET

COPPER BLOCK

WATER LINES

Fig. 5. Internal-beam target and holder.

internal beam was suggested, since currents up to 100 μa, or more, could readily be obtained.

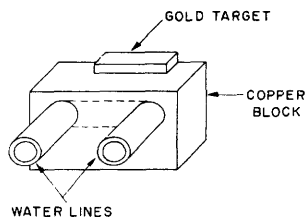A target holder was designed to hold an ingot of gold $1 \times \frac{1}{4} \times \frac{1}{8}$ in. in the internal beam. The holder was made from a block of solid copper for optimum cooling, with copper tubes of $\frac{1}{2}$-in. diameter serving as water inlet and outlet pipes (Fig. 5). The holder was designed so that after bombardment the water lines could be unsoldered from the stubs, and the holder and target slipped into the vise of a lead-lined milling machine. The planned procedure was to remove the top 0.010–0.020 in. of the gold target and treat these chips just as the gold foil was handled. The milling machine had been built so that all chips removed from the target would be deposited in a tray that could be manipulated by means of extension rods.

In the only internal beam run, the gold target accidentally fell from the face of the holder while the water

202
200
204 199
199 201 198 201 201
$\nu \rightarrow$ mK

Hg I   4047 A

202
199 200 198 199
201 201 201
204

Hg I   2537 A

FIG. 6. Hyperfine structure of the 2537-A and 4047-A lines of Hg. Upper parts of the photographs are natural mercury; lower parts are the decay products (found in the lamp) of the internal beam run. The right-hand sides identify the lines.

lines were being unsoldered. After most of the solder was mechanically removed, the entire gold ingot was treated just as the gold foil described above. Fortunately, residual solder did not interfere with the remainder of the experiment. In this run the cyclotron was operated at 50 μa for 6 hr. Ten millicuries, or approximately 0.001 μg of radio-thallium was extracted from the 10-g gold target. This proved to be enough thallium to obtain the desired results.

Another interesting feature of the internal beam run was the character of the hyperfine-structure patterns of the two mercury lines at 2537 and 4047 A. Spectrograms of these lines taken after the lamp that was made from this bombardment was allowed to cool for three weeks show the presence of only Hg[199] and Hg[200] (see Fig. 6). These two isotopes are just the decay products of Tl[199] and Tl[200]. In other words, there was no natural mercury contamination, in spite of the fact that the lamp was prepared on a mercury-pump vacuum system.

### 2. Tl[201] (3 day), Tl[202] (12 day), and Tl[204] (4 yr)

These three isotopes of thallium were produced in the M.I.T. cyclotron by deuteron bombardment of mercury in the external beam. The deuteron beam is stronger and more reliable than the alpha beam and currents of approximately 30 μa of 15-Mev deuterons were used to irradiate the target for periods of 12–20 hr in these bombardments. Many reactions occurred. The reaction products for which some evidence was found—spectroscopic, half-life count of the target or gamma-ray spectrum—are the following: Tl[200], Tl[201], Tl[202], Tl[203] (relatively small quantity), Tl[204], and Tl[205].

Because all these isotopes are produced simultaneously, an accidental overlap of components masked the presence of Tl[204] in the first spectrograms. This problem was overcome by performing the experiment twice: the first time, the spectra of Tl[201] and Tl[202] were obtained; the second time, the spectrum of Tl[204] was obtained by waiting one week for the Tl[201] to die away before taking the spectrograms.

A standard external beam target holder was modified[12] to hold liquid mercury during bombardment. One face of the holder was milled flat, and a series of holes, slightly less than $\frac{1}{8}$ in. in diameter, were drilled approximately $\frac{1}{8}$ in. apart and $\frac{1}{16}$ in. deep into this flat face. The holes were filled with mercury and covered with an aluminum foil that was 0.0015 in. thick. Then the target area was surrounded by a Teflon gasket, 0.002 in. thick. A second aluminum foil, similar to the first, covered the gasket, and the aluminum–Teflon sandwich was held in place on the holder by means of an aluminum plate $\frac{1}{16}$ in. thick, bolted to the face of the holder and louvered to permit passage of the beam.
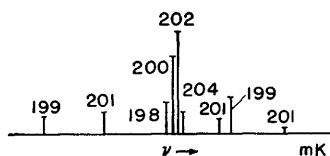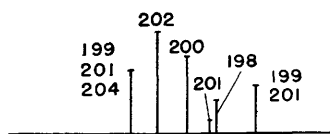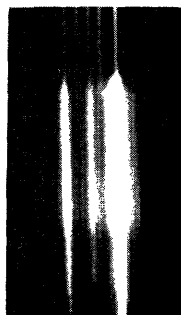
The deuteron beam burned the inner foil in places and the mercury was discolored; but the outer foil was completely undamaged and held the mercury in place.

After bombardment, the aluminum foils were cut away by means of a knife blade attached to a 3-ft rod. The mercury was removed from the target and deposited in a Vycor test tube in a one-step operation. An aspirator assembly was built to perform this operation (see Fig. 7). The test tube containing the mercury was sealed into a cell on the vacuum system, just as the gold was in the previous runs. The cell was baked at 200–250°C for 2 or more hours while it was being pumped until all of the mercury had been driven from the cell. The rest of the procedure for preparing the lamp was identical to the earlier runs, but more care was required to ensure that most of the mercury was removed from the lamp before sealing it off the system. In fact, some mercury remained in the lamps in both of these runs, but the contamination was not severe.

III. RESULTS AND DISCUSSION

A. Hyperfine Structure Measurements and Nuclear Moments

The hyperfine structure separations $\Delta\sigma$, of the ground state $6p\,^2P_{\frac{1}{2}}$ and of the excited state $7s\,^2S_{\frac{1}{2}}$ have been measured for the odd-even isotopes of thallium of mass numbers 199 and 201. Values of $\Delta\sigma(^2P_{\frac{3}{2}})$ for these isotopes have been estimated. Three spectrograms were obtained of the ultraviolet line of Tl^{199}, and two of its green line. Photographs of two of these spectrograms, together with energy-level diagrams are given in Figs. 8 and 9. The intervals between components of the same isotope on the plates of the ultraviolet lines are directly
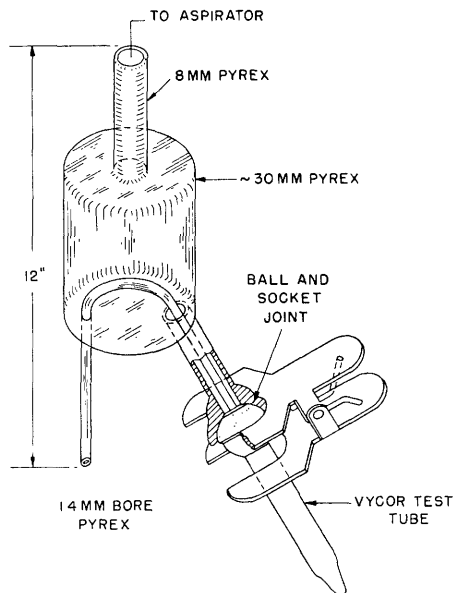
Fig. 7. Aspirator assembly for transferring radioactive mercury from target holder to vacuum system.

proportional to the hfs splittings of the $^2P_{\frac{1}{2}}$ and the $^2S_{\frac{1}{2}}$ states. The single interval per odd-even isotope on plates of the green line is proportional to $\Delta\sigma(^2S_{\frac{1}{2}})$ $-\Delta\sigma(^2P_{\frac{1}{2}})$. These interval proportionalities are illustrated in Fig. 2. If 17 mK is chosen as an estimate of the $P_{\frac{1}{2}}$ splitting and is used to obtain the splitting of the S state from the green line, this value agrees well with that obtained from the ultraviolet line.

The values of $\Delta\sigma(^2P_{\frac{1}{2}})$ and $\Delta\sigma(^2S_{\frac{1}{2}})$ of Tl^{201} could not be obtained directly from the ultraviolet line because

Fig. 8. Hyperfine structure of the 3776-A line of Tl^{199} and Tl^{200}.

FIG. 9. Hyperfine structure of the 5350-A line of Tl[199] and Tl[200].

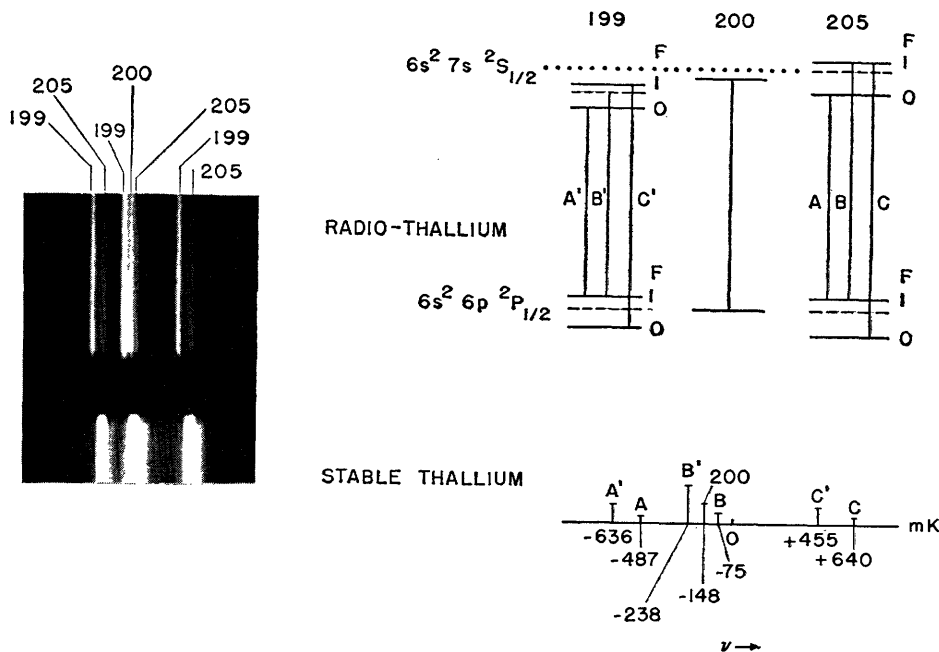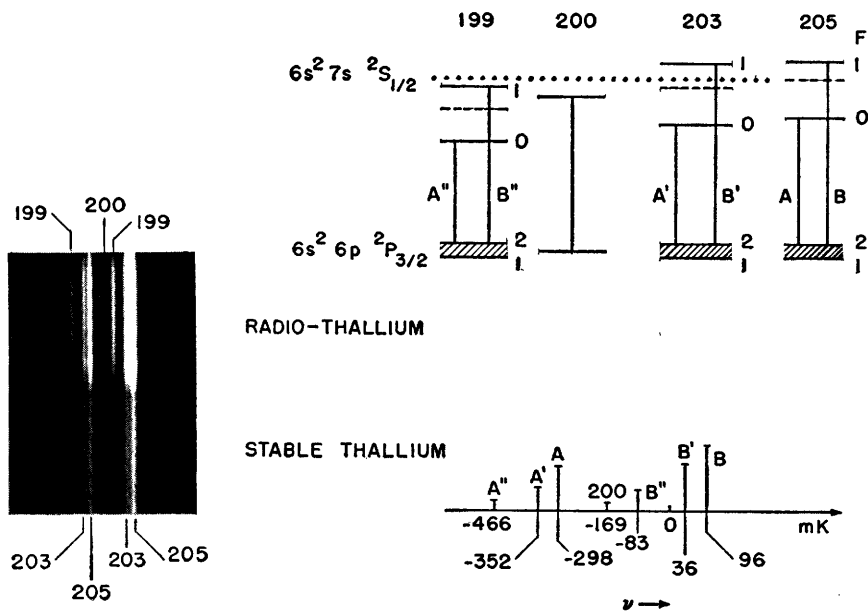of overlap of its central component with Tl[200] (see Fig. 10). However, the total spacing $\Delta\sigma(^2P_{\frac{3}{2}}+^2S_{\frac{1}{2}})$ could be measured. Then the ratio of the S-state splitting to the total spacing was averaged for Tl[199], Tl[203], and Tl[205]. This average ratio was multiplied by the total spacing in Tl[201] in order to obtain $\Delta\sigma(^2S_{\frac{1}{2}})$ and from this, the ground-state splitting. Again, this value for the $^2S_{\frac{1}{2}}$ splitting was in good agreement with that obtained from the green line by taking 17 mK for $\Delta\sigma(^2P_{\frac{3}{2}})$. Fig. 10 also shows the Tl[202] spectrum, and it can be seen again in Fig. 11 together with that of Tl[204].

These results are given in Table I. The half-life or the natural abundance is listed in the column under $T_{\frac{1}{2}}$. The spins, listed in the column under I, had all been determined previously, and were verified for Tl[199] and Tl[201] in this study. The values of the hfs splittings are in millikaysers. The magnetic moments, μ^{exp}, are calculated from the observed splittings by comparison with the stable thallium hfs and the moments obtained from nuclear magnetic-resonance experiments. The references a–h of Table I were not measured by the authors but are included for completeness.
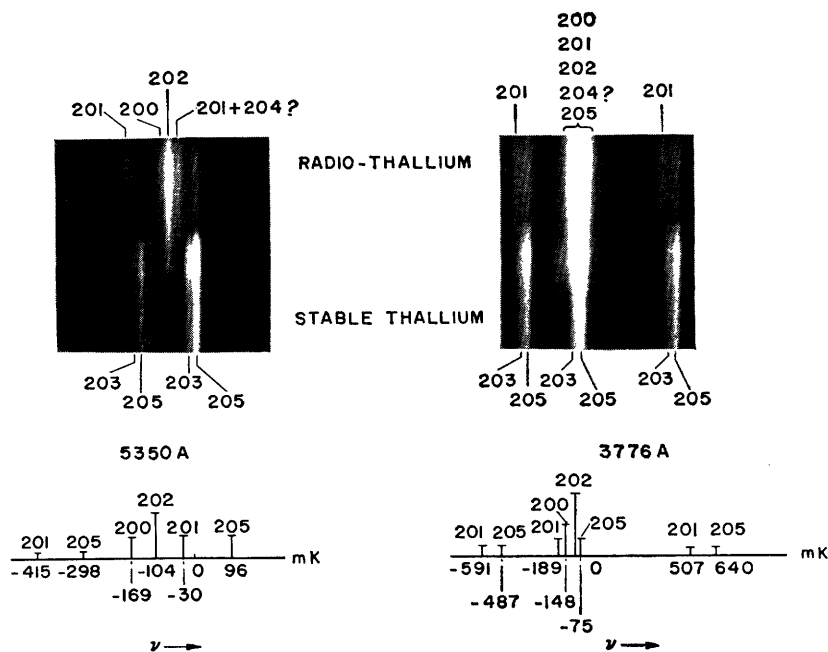

FIG. 10. Hyperfine structure of Tl[201] and Tl[202].

Upper limits have been placed on the splittings in the ground states of the odd–odd isotopes. These limits were inferred from the observed linewidths. The corresponding upper limits on the magnetic moments are also listed in Table I.

The uncertainty associated with each of the hfs splittings measured in this work is ± 4 mK. Standard deviations were calculated for a few of the intervals on the spectrograms. These deviations were all close to 4 mK; therefore the error quoted above is approximately one standard deviation.

The presence of a weak unresolved third component in the hfs pattern of the green line (Fig. 2) was expected to produce a shift in the measured interval. However, a graphical summation of the two components, with relative intensities 5:1, a Doppler width of 30 mK, and a 17-mK separation of line centers, showed a shift in the maximum density of the resulting line of less than 2 mK. Since the shift was less than half of the error limit quoted and the results from the green and ultraviolet lines showed agreement, it was neglected.

Although M.I.T. grating No. 97 is, in fact, excellent, we thought that it would be worth while to consider systematic errors that may arise from distortion of the ideal diffraction pattern as discussed recently by G. W. Stroke.[13] His calculations of the diffraction pattern produced by this grating showed the presence of a weak satellite on one side of the main peak. We have taken the convolution of the actual instrument function for monochromatic light with a Doppler-broadened line of 30-mK width for a wavelength of 5461 A. The resultant shift in the peak is approximately 1 mK, and would appear in all lines measured with a linear detector. The nonlinearity of the photographic plate could therefore lead to errors of this amount for lines of vastly different intensities. We have estimated the error introduced in the separation of two Doppler-broadened lines, which is caused by their nearness to each other, and find this effect to be also well within our quoted error limits.

The last two columns of Table I list values of the magnetic moments for the odd-even isotopes calculated by using the configuration-mixing theory of Arima and Horie.[14] The following contributing nucleon configurations were assumed for calculating $\mu^{\text{th}}{}_I$: (a) proton, $(1h_{11/2})^{12}s_{1/2}$; (b) neutron, $(3p_{3/2})^4(1i_{13/2})^{N-110}$, where N is the number of neutrons in the isotope under consideration. For $\mu^{\text{th}}{}_{II}$, we assume the less likely possibility—that the p-neutron shells are completely filled before the i shell. The Schmidt limit for an odd $s_{\frac12}$ proton is 2.79 nm. Although agreement between experiment and theory is not very good, the important observation is that configuration mixing can account for large deviations from the single-particle model.

We note that for all the odd-Z odd-N thallium iso-
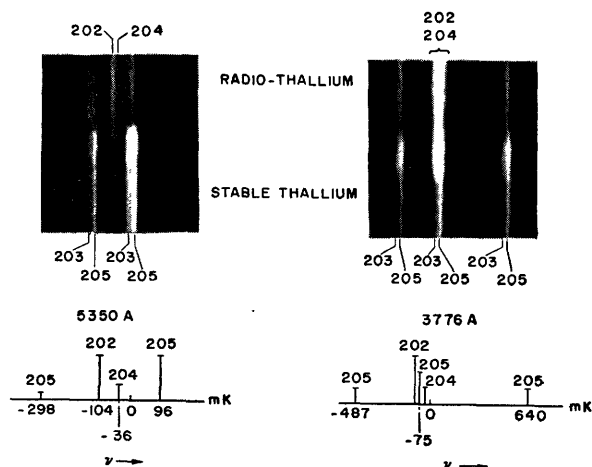
FIG. 11. Hyperfine structure of Tl^{204}.

topes the magnetic moments are very small (0.089 nm for Tl^{204}). A picture in which the $3s_{\frac12}$ proton is coupled with the $3p_{\frac32}$ neutron, and in which the empirical proton g factor obtained from the odd-A thallium moments and the neutron g factor from $\mu(\text{Hg}^{201})$ are used, would give a moment of approximately 1 nm. Configuration mixing[15] does not give a better result. De-Shalit[16] suggests the following coupling scheme. We couple a $3p_{\frac32}$ neutron (using the Hg^{199} g factor) to a 2+core ($g \approx 0.4$) to form an angular momentum of $\frac52$. This is then coupled to the $3s_{\frac12}$ proton. With this scheme good agreement can be obtained with the experimental value of $\mu(\text{Tl}^{204})$ even when admixtures from other states are neglected. The lack of beta decay of Hg^{203} to the ground state of Tl^{203}, which has spin $\frac12$, would lend support to the existence of such a spin-$\frac52$ state. On a purely single-

TABLE I. Hyperfine-structure splittings and magnetic moments.

A	$T_{\frac12}{}^a$	I	$^2P_{\frac12}$	$^2P_{\frac32}$	$^2S_{\frac12}$	μ^{exp}	$\mu^{\text{th}}{}_I$	$\mu^{\text{th}}{}_{II}$
			$\Delta\sigma$(mK)			Magnetic moments (nm)		
199	7.4h	$\frac12$^{b}	693	17	398	1.57	1.38	1.63
200	27h	2^c	<40			$\lvert\mu\rvert \leqslant 0.15$		
201	3d	$\frac12$^{c}	696	17	402	1.58	1.36	1.61
202	12d	2^c	<40			$\lvert\mu\rvert \leqslant 0.15$		
203	30%	$\frac12$^{d}	704.2^e	17.5^f	405^i	1.60^g	1.33	1.58
204	4y	2^b	24.4^h			± 0.089^{b}		
205	70%	$\frac12$^{d}	711.0^e	17.7^f	412^i	1.61^g	1.31	1.56

a D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).
b G. O. Brink, J. C. Hubbs, W. A. Nierenberg, and J. L. Worcester, Phys. Rev. **107**, 189 (1957).
c L. L. Marino, Report UCRL-8721, Radiation Laboratory, University of California, April, 1959.
d P. Brix and H. Kopfermann in *Zahlenwerte und Funktionen*, *I/5*, edited by H. H. Landolt and R. Börnstein (Springer Verlag, Berlin, 1952).
e A. Lurio and A. G. Prodell, Phys. Rev. **101**, 79 (1956).
f G. Gould, Phys. Rev. **101**, 1828 (1956).
g H. E. Walchli, Report ORNL-1469, Oak Ridge National Laboratory, April 1953.
h J. Winocur, R. Marrus, J. C. Hubbs, and A. Cabezas, Bull. Am. Phys. Soc. **3**, 415 (1958).
i R. J. Hull and H. H. Stroke, J. Opt. Soc. Am. **49**, 1088 (1959).

13 G. W. Stroke, Rev. opt. **39**, 291 (1960).
14 A. Arima and H. Horie, Progr. Theoret. Phys. (Kyoto) **12**, 623 (1954); Phys. Rev. **99**, 778 (1955).
15 H. Noya, A. Arima, and H. Horie, Progr. Theoret. Phys. (Kyoto) Suppl. **8**, 33 (1958).
16 A. de-Shalit, Massachusetts Institute of Technology, January, 1961 (private communication).

particle basis this spin would be somewhat unlikely in this neutron region, and, also, it would not give satisfactory results for $\mu(Tl^{204})$.

B. Isotope Shifts

The results of the isotope-shift measurements are given in Table II. The shifts are given in millikaysers. All shifts were measured with respect to Tl^{205}. The uncertainties are, again, ±4 mK.

The isotope shifts were obtained by calculating the centroid of the hfs pattern of the odd-even isotopes

TABLE II. Isotope-shift data.

Mass number	Ultraviolet line (mK)	Green line (mK)	Relative shifts Thallium	Relative shifts Mercury
198	...	...	...	−0.92
199	−164	−177	3.00	−0.80
200	−148	−169	2.86	0.00
201	−115	−124	2.10	0.30
202	...	−104	1.76	1.00
203	−59	−59	1.00	...
204	...	−36	0.61	1.99
205	0	0	0	...

with the use of the interval rule. In calculating shifts from the ultraviolet line the ground states of the isotopes were set at equal energies. The metastable states were set equal when the green line was used. When both values were obtained, they usually differed by more than the limits of error. This indicates that at least two of the three states participate in the shift. Odintsov[17] found recently that the $7^2S_{\frac{1}{2}}$ state has a positive shift of 14.9 mK, while the $6\,^2P_{\frac{3}{2}}$ state shows a negative shift of 44.0 mK; both are referred to the $10\,^2P_{\frac{3}{2}}$ state. These figures are for the thallium 203–205 shift. This is in good agreement with the present work for the isotope shift in the green line.

It is possible to explain the difference in shifts in the ultraviolet and the green lines. A rough calculation for electrons with $l>0$, given by Breit,[18] shows that the $5d^{10}6s^26p\,^2P_{\frac{1}{2}}$ term ought to have a contribution of approximately 34 mK to the isotope shift from the $6p$ electron. The shift of the $^2P_{\frac{3}{2}}$ term should be approximately 5000 times less, contributing a completely negligible amount to the $5d^{10}6s^2$ shift. Also, the $p_{\frac{1}{2}}$ electron is expected to produce a greater screening effect than the $p_{\frac{3}{2}}$ electron. Thus the final result of this positive shift and the screening effect could be the difference in shifts for the two P levels. The main difficulty in these estimates is the calculation of the screening effects because very accurate electronic wave functions are required to make proper calculations. Screening effects can also account for the negative shifts. Detailed discussions of these screening effects are given by Kopfermann[6] and by Crawford and Schawlow.[7]
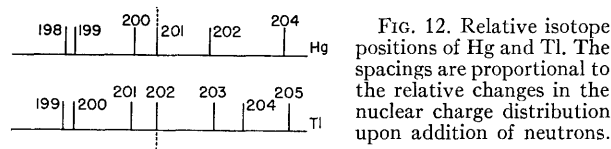

FIG. 12. Relative isotope positions of Hg and Tl. The spacings are proportional to the relative changes in the nuclear charge distribution upon addition of neutrons.

Values of the isotope shift could not be obtained in the ultraviolet line for two of the isotopes because of overlap of components with Tl^{205}, which was present in varying quantities in every run.

The fourth column of Table II gives the result of dividing each of the shifts in the green line of thallium by the 203–205 shift. The green line was used because shifts were available for each of the isotopes. These relative shifts are also given in Table II for the isotopes of mercury with the same mass numbers as the thallium isotopes. These values are those given by Kopfermann.[19] When the relative shifts of both elements are compared with respect to neutron number, there is a striking similarity. Figure 12 shows this comparison; the isotopes with $N=121$ are arbitrarily set on the same vertical line. This procedure indicates that the relative isotope shifts are roughly independent of the proton numbers, but the shifts show a marked dependence on neutron number. Kuhn and his co-workers[1–3] have made a similar comparison of the relative isotope shift with neutron number. They have worked with stable, even-even isotopes from $Z=46$ to $Z=50$ (Mo,Pd,Cd,Sn). From their experimental results they also draw the conclusion that there is a definite dependence of relative isotope shift on neutron number, but they infer in addition a dependence on proton number.[3] In the present case we can account for the great similarity of the relative isotope shifts in Hg and Tl and their apparent independence of the presence or absence of the odd proton. The proton that we add to Hg to produce Tl is in an $s_{\frac{1}{2}}$ state. We expect therefore the additional charge to be distributed with spherical symmetry over the A nucleons and therefore have a relative effect on the isotope shift which is too small to be detected here.[20] It will therefore be of interest to obtain a comparison of Hg and Au isotope shifts: in Au the odd proton no longer has a spherically symmetrical distribution but is in a $d_{\frac{3}{2}}$ state and could therefore lead to greater distortions through a polarization of the core.

The present work gives the first instance in which the isotope shift has been observed for heavy odd-odd nuclei. The well-known odd-even staggering effect,[21] which is present for the stable isotopes of odd mass number, is also present in the odd-odd isotopes of thallium: the effect is evidently mainly a function of neutron number. For thallium, the centers of gravity of the electronic-term positions of the even-numbered isotopes lie closer to the next lighter isotope.

[17] A. I. Odintsov, Optics and Spectroscopy 9, 75 (1960).
[18] G. Breit, Revs. Modern Phys. 30, 507 (1958).

[19] See reference 6, p. 181.
[20] We are indebted to Dr. V. Jaccarino, Bell Telephone Laboratories, for a valuable discussion which led to this explanation.
[21] See reference 6, p. 436.

Various explanations of the odd-even staggering effect have been suggested by Wilets, Hill, and Ford,[22] and by Bodmer.[23] A possible explanation, suggested by Brown,[24] is that two particles added to a parent nucleus can already be coupled to an angular momentum 2 state and therefore interact more strongly with the "core" than a single nucleon which would first have to be coupled by a Clebsch-Gordan coefficient to the parent nucleons before producing such "core" excitations. This would then result in the observed effect that the relative increase in the nuclear charge distribution upon the addition of an odd neutron is less than half of the increase produced by a neutron pair. A review of theoretical work on isotope shift has been given by Breit.[18]

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APPENDIX

Preparation of Electrodeless-Discharge Lamps

The main difficulty in our experiment was the production of a lamp that would last at least 2 hr, that is, a lamp that would emit the spectrum of thallium for this time. After several failures of the experiment with radio-thallium, an investigation of the lifetimes of electrodeless-discharge lamps containing minute quantities of material was started. For these experiments natural thallium rather than radio-thallium was used. A standard procedure was developed for making the lamps.

A solution containing a known concentration of thallium was prepared in the form of thallium sulfate. A drop of solution containing 0.01 μg of thallium was placed in a chemically cleaned quartz or Vycor tube. This cell was warmed under a heat lamp to evaporate the liquid slowly and leave a residue of thallium. All of the lamps started with this quantity of thallium and this kind of operation.

After the solutions were dried the cell was sealed onto the vacuum system. The system was baked at 200°C for 1 hr and then liquid nitrogen was placed on the traps. The section of quartz tubing which was to become the lamp was outgassed by various means to determine the best method. These methods included gentle heating ($\sim$500°C) for 2 hr with a gas-air flame, moderate heating ($\sim$1000°C) for 2 hr with an electric

furnace, and intense heating ($\sim$1500°C) for a few minutes with a gas-oxygen flame. Each method seemed equally satisfactory, so that the last and simplest method was adopted. In any event, there seemed to be no direct dependence of lamp lifetime on the cleanliness of the walls, but there were two effects to be considered: (1) a really dirty lamp will produce a more or less continuous background that may mask the desired spectrum, and (2) a dirty lamp is more difficult to keep lit, especially when it is heated, probably because of pressure build-up.

An rf discharge was run in the lamp before it was sealed off the system. If the spectrum appeared to contain a high background or many impurities when viewed in a hand spectroscope, the discharge was run at full power ($\sim$50 w) for a few minutes, the discharge shut off, the lamp evacuated, and refilled with argon. This flushing procedure was repeated until the discharge appeared clean. Usually two or three flushings were sufficient.

There was one observation made on the behavior of the radio-thallium lamps which served as a guide for some of the following experiments: Quite often a brand new lamp would show the characteristic thallium green line and a dense impurity spectrum including CO bands, when the lamp was first excited and gently heated. After 10–15 minutes of operation the impurity spectrum would disappear quite suddenly and, soon thereafter, the thallium spectrum would also disappear. This led to attempts to introduce into the lamp only known impurities with reasonably simple spectra. To eliminate unknown impurities, the vacuum system was baked before producing a trial lamp at 350°–400°C for 2 hr.

Gallium and indium (chemically similar to thallium) were introduced into the cell in solution for different lamps as the first impurities. Approximately 1 μg of indium had the most noticeable effect, increasing the lifetime of the lamp from 10 minutes to approximately 1.5 hr. Addition of greater or lesser amounts of indium by factors of 10 did not change this figure appreciably. Gallium seemed to have no effect.

Lamps of various sizes were made. The largest showed no thallium spectrum at all in the Hilger spectrometer, probably because of the extremely low density of thallium atoms. The smallest lamp (1-mm diameter, 3 cm length) showed no increase in lifetime over the standard lamp (3 mm diameter, 5 cm length).

Various pressures of filler gas were used. Although there was no difference in lifetime, lamps filled with argon at a pressure of just 1 mm Hg gave least trouble in starting and staying lit when heated.

Microwave excitation at approximately 3000 Mc produced a more intense spectrum than rf excitation at 50 Mc for a short time; but there was no increase in lamp life.

Various methods were tried to lower the electron temperature in the lamp. The object was to prevent

[22] L. Wilets, D. L. Hill, and K. W. Ford, Phys. Rev. **91**, 1488 (1953).
[23] A. R. Bodmer, Proc. Phys. Soc. (London) **A67**, 622 (1954).
[24] G. E. Brown, Nordita, Copenhagen, August, 1961 (private communication).

the thallium atoms from becoming ionized, in the hope that if the atoms were driven into the walls only because of thermal agitation, they could be driven back into the discharge by heating the walls. Nitrogen, with many bands in the visible region, and xenon, which has the lowest ionization potential of the noble gases, were used in various combinations with argon. Sodium and lithium, with resonance lines involving only a few volts of energy, were used for this purpose along with indium in the lamp. No improvement from either the gases or the alkali-metals resulted.

The technique that we finally adopted was to dope each lamp with 1 μg of indium and fill the lamp with the maximum number of atoms that could be produced.