DIPOLE AND QUADRUPOLE MOMENTS OF THE ISOMERIC Hg\textsuperscript{197} NUCLEUS; ISOMERIC ISOTOPE SHIFT

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The hyperfine structures of five optical lines of 65-hour radioactive Hg$^{199}$ and 25-hour isomeric Hg$^{201}$ were measured. Radioactive mercury produced by cyclotron bombardment and excited in electrodeless discharge tubes containing as few as 5X10$^{12}$ atoms, free of natural mercury, gave adequate light intensities. The analysis of the hyperfine structure confirmed the magnetic dipole moment and isotope shift of Hg$^{199}$ in its ground state ($I=\frac{1}{2}$), and gave the following values for Hg$^{201}$ (based on $I=13/2$): $\mu_H^{199} = -1.04\pm0.01$ nm; $Q_{blend} = (1.5\pm0.3) \times 10^{-6}$ cm$^2$; isotope shift displacement (from Hg$^{199}$ in the 2537 A line) $+70\pm7\times10^{-8}$ cm$^{-1}$. Thus an isomeric shift of atomic energy levels resulting from the excitation of the nucleus from its ground state $I=\frac{1}{2}$ to the isomeric state $I=13/2$ is observed. In the 2537 A line of mercury this shift amounts to $-21\pm6\times10^{-8}$ cm$^{-1}$.

**INTRODUCTION**

We report here the results of a spectroscopic investigation of several optical lines of the radioactive Hg$^{199}$* isotope; the nucleus of this atom is in a 25-hour excited isomeric state ($I=13/2$). The existence of this nuclear isomeric state has been known for several years and has been discussed in various papers.$^{1,2}$ The nuclear energy level scheme for Hg$^{199}$* was determined by analyzing the spectrum of the internal conversion electrons, combined with coincidence$^3$ and angular correlation$^4$ measurements; it is shown in Fig. 1. The proposed spins for the 25-hour isomeric and 65-hour ground states ($I=13/2$ and $I=1/2$, respectively) are in agreement with the shell model$^5$ and were confirmed by our results.

In an early paper$^6$ several new components in the 2537 A resonance line and the 4047 A line were reported. The analysis of the Hg$^{199}$ components gave $\mu_{199}/\mu_{199} = A_{197}/A_{199} = 1.04\pm0.03$ for the ratio of the moments. Recently,$^7$ it became possible to establish a double (paramagnetic) resonance in the $F=3/2$ level of the $3^P_1$ state of Hg$^{199}$ and to obtain $A_{197}/A_{199} = 1.045 \pm 0.002$. Further, the combination of the double-resonance method with magneto-optic scanning$^8$ gives for the isotope shift (from Hg$^{199}$) $91\pm4$ mK [where 1 mK (millikayser) $= 10^{-6}$ cm$^{-1}$].

The energy of an hfs component is given by

$$W_I = W_J + AC + B - C(J+1-J(J+1)),$$

with

$$C = F(F+1) - (I+1) = J(J+1),$$

where $F$ is the total angular momentum of the component, $I$ is the nuclear spin, and $J$ is the angular momentum of the atomic level; $A$ and $B$ are the magnetic dipole and electric quadrupole interaction constants, and $W_J$ is the energy of the center of gravity of the hfs multiplet.

The 2537 A, 4047 A, 4078 A, 4358 A, and 5461 A lines were investigated; the corresponding energy levels are shown in Fig. 2. The Hg$^{199}$ components were immediately identified because the structure of the lines could be deduced from the knowledge of the $3^P_1$ interaction constant.$^8,9$ However, some blending of Hg$^{199}$* and Hg$^{201}$ components exists, but it is possible to resolve them by combining the data from several lines. For the interaction constants of natural mercury and the structure of the mercury hyperfine structure we used the results of Blaise and Chantrel.$^9$

**EXPERIMENTAL PROCEDURE**

The radioactive samples were prepared in the MIT cyclotron by the Au$^{197}(d,2n)$Hg$^{199}$ reaction, and amounts of as much as 15 millicuries were easily extracted from the gold target (approximately 2X10$^{14}$ atoms). Usually the full energy of the beam (15.2 Mev) and a thick target (0.005 inch) were used, so that the Au$^{197}(d,3n)$Hg$^{196}$ and Au$^{197}(d,n)$Hg$^{198}$ reactions also occurred. These occurrences were confirmed by the spectroscopic work on the 2537 A line, from which the following estimate of

the relative constitution of the samples was deduced:
\[ \text{Hg}^{197*} \approx 17\% ; \text{Hg}^{197} \approx 54\% ; \text{Hg}^{196} \approx 17\% ; \text{Hg}^{198} \approx 12\% . \]

The main difficulty with previous spectroscopic investigations was contamination of the sample by natural mercury. We were able to overcome this disadvantage by moderately heating the target in vacuum after bombardment. This was necessary because it was impossible to prevent the adsorption of natural mercury on the surface of the gold target during bombardment. A detailed description of the sample preparation technique is given by Melissinos.\(^{10}\)

The source was an air-cooled electrodeless discharge. It consisted of a 7-mm Vycor tube containing from \(10^{12}\) to \(2\times10^{14}\) atoms of radioactive mercury and argon at a pressure of several millimeters (estimated 10 mm); the discharge tube was easily excited with a 60-Mc/sec oscillator but was operated at minimum power, so that only one-fifth to one-tenth of the length of the tube was filled with a discharge—with the double advantage of giving narrow lines and reducing the clean-up rate. After operating the tube for approximately 15 minutes the mercury spectrum would disappear and, instead, strong molecular bands appeared. These were attributed to impurities from organic materials present in the vacuum system used in filling the tube. However, a thorough flaming of the tube for a minute or so, re-established the mercury spectrum. The clean-up rate depended upon the amount of radioactive mercury in the tube, but with a little experience in preparation and operation it was possible to run tubes with \(3\times10^{13}\) atoms for one hour before flaming was needed.

The spectrograph consisted of one of Harrison's\(^{11}\) 10-inch diffraction gratings used in autocollimation in a mirror monochromator with a focal length of 40 feet.

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\(^{10}\) A. Melissinos, Ph.D. thesis, Department of Physics, Massachusetts Institute of Technology (1958, unpublished).

Because of the accurate groove spacing and intense blaze, this grating can be used at high angles of incidence and diffraction, with little loss of light or resolution. It has a groove spacing of 300/mm and throws about 35% of the light (as compared to an aluminized mirror of the same aperture) into a very small range of angles near 63°. Most of the spectrograms were taken near this angle, with 2537 Å appearing at 66° in the 24th order, with a plate factor of 700 mK/mm (0.045 Å/mm). The plate factors at each wavelength were calculated from the known hyperfine structure of natural mercury. The measured resolving limit at 5461 Å closely approaches its theoretical value of 22 mK. Exposure times varied from a few minutes for 2537 Å to four hours for 4078 Å, on Kodak types 103 a-o and F spectroscopic plates. In each case a rotating sector was used to give varied exposure times—wide slits being used in some cases to increase intensities.

Four successful runs were made, as well as a preliminary interferometric investigation. The decay of various hfs components was checked spectroscopically and confirmed our assignments; it also indicated that the only stable isotopes contained in the tubes were Hg²⁰⁷ and Hg²⁰⁹ (and a 2–5% natural mercury contamination). As an example we give in Fig. 3 four exposures of the 2537 Å line with sources at different stages of decay. Three days after bombardment the Hg²⁰⁷ component nearly vanishes; after 10 days it has completely disappeared and Hg²⁰⁹ has become weak. After 30 days, both have disappeared, and only the stable isotopes are left.

The radioactivity of the discharge tubes was checked with a 256-channel analyzer for a normal γ spectrum, and indicated the presence of Hg²⁰⁷* and Hg²⁰⁹. (See Fig. 1.)

**EXPERIMENTAL RESULTS**

Our results are summarized in Figs. 3–7. The position of each component is given in millikaysers; usually from Hg²⁰⁹, which was placed on the plates for comparison. The energy level spacings were derived from the respective lines unless they are in parentheses or brackets. (Parentheses indicate that this particular spacing was obtained from some other line; brackets indicate estimated values.)

**2537 Å (6s² ¹S₀–6s6p ³P₁)**

Several successful exposures of this line were taken (Fig. 3). Component b was obtained from an exposure with a special source which did not contain Hg²⁰⁹. We obtained this source by reducing the deuteron beam energy to 11.2 Mev and using only a 0.002-inch gold target. Component a was never resolved because it overlaps the Hg²⁰⁷, F=1/2 line, but its position was obtained from the 4358 Å data.

Discussion of the analysis of this line will be presented after the 4358 Å data have been considered, since these data give the most reliable values of the spacings between the levels of the ³P₁ state.

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13 We are greatly indebted to Professor L. C. Bradley III, who provided the interferometer and took the exposures.
4047 A (6s6p 3P0 − 6s7s 3S1)

The positions of the components in this line were obtained from three exposures; its structure was confirmed from more than four exposures. The structure of the line and the energy level scheme are given in Fig. 4.

Only two components of Hg197*, b and c, are resolved. However, since we know that no other isotopes are present and that all lines are correctly accounted for, we deduce that b and c belong to an hfs triplet. The third component of this triplet must be located in the negative wave-number region in order to give a reasonable position for the center of gravity. We assumed that the missing component a overlaps A.

Further, since the 3S1 state has zero quadrupole interaction, the interval rule gives us a value for the nuclear spin. We have the relation

\[(I+1)/I = (852±3)/(739±3),\]

which yields

\[I = (13.1±0.6)/2.\]

Using the value 13/2 for the spin, we readily obtain the magnetic dipole interaction constant for the 3S1 state

\[A_{197+}(3S_1) = -113.6±0.5 \text{ mK}.\]

With the identification of the 3S1 energy levels we are then able to proceed to the analysis of the 4358 A and 5461 A lines.

4078 A (6s6p 3P1 − 6s7s 3S0)

A 4-hour exposure of this line gave an extremely satisfactory picture; its structure and the energy levels are given in Fig. 5. Unavoidably, the components were quite broad because (a) the discharge tube was operated at a high power level, (b) a wide slit was used, and (c) the exposure was very long.

Components A, B, and 196+198 were resolved in the interferometric work as well. Component b was not completely resolved in either. Component a is quite strong and well defined in the grating exposure. The third component, c, of Hg197*, overlaps B, their separation being only 25 mK.

Since component b is not completely resolved, it was not used for the determination of the \(F = 11/2 - F = 13/2\) spacing of the 3P1 state. Component a combined with the 2537 A data can be used to give the "isomeric shift." Any difference between the spacings \(c - B = 297\) mK in the 2537 A line and \(A - a = 310\) mK in the 4078 A line, results from isotope shift in the lines and represents the difference of the isomeric isotope shifts in the 2537 A and 4078 A lines. Further, since the isotope shift in the 4078 A line is known to be one-sixth of the shift in the 2537 A line we can obtain the isomeric shift in the 2537 A line.

\[(A - a) - (c - B) = \text{isotope shift (4078) - isotope shift (2537)} = -13 \text{ mK. Thus}

\[\text{isomeric shift (2537 A)} = -16 \text{ mK.}\]

The advantage of this calculation is that it is independent of any value of the interaction constants that are needed to find the actual centers of gravity; further, all four components are free from overlapping. The value thus obtained is in good agreement with the straightforward calculation of the centers of gravity of Hg197 and Hg197* in the 2537 A line, which yields \(-21±6\) mK. (See the discussion at the end of this section.)

Note that the center of gravity of Hg197* given in
Fig. 5 was obtained by using the best values for the interaction constants of the $^3P_1$ state.

**4358 Å ($6s6p\ ^3P_1-6s7s\ ^3S_1$)**

Two good exposures of this line were obtained—a weak one and a strong one—and they both reveal ten well-defined components (Fig. 6). Components $A$, $B$, $C$, $D$ belong to Hg$^{197}$, and except for $B$ they check correctly with the known intervals in the $^3S_1$ and $^3P_1$ states. The even-isotope component, labeled “196+198,” appears between $B$ and $C$. The remaining five components belong to Hg$^{197}$. Altogether there are seven Hg$^{197}$ transitions, but $d$ cannot be observed because of extremely low relative intensity, and $b$ blends with $B$. To this blending we attribute the shift of the $B$ component which we expected at $-263$ mK.

Accepting the intervals of $739$ mK and $852$ mK for the $^3S_1$ state, we deduce that components $a$ and $g$ are correct and that the $(c-f)$ difference has a discrepancy of $14$ mK from the expected value. Therefore, the lower interval of the $^3P_1$ state was obtained from components $e$ and $a$ (or $e$ and $g$), while the upper one is the average of the values given by $(g$ and $f)$ and $(a$ and $c)$.

**5461 Å ($6s6p\ ^3P_2-6s7s\ ^3S_1$)**

For this line, eight components were resolved. Three of them, $A$, $C$, and the central one, correspond to the three Hg$^{197}$ transitions. The $(A-C)$ difference checks...
correctly with the hfs interval of the $^3S_1$ state. Because the components from Hg$^{196}$ and Hg$^{198}$ overlap component $B$ of Hg$^{197}$, they give rise to an extremely broad line, and therefore the position of $B$ is not reliable. For this reason we do not give a value for the interaction constant of Hg$^{197}$ in the $^3P_2$ state.

The structure of the line and the energy level scheme are given in Fig. 7. The five components—$a$, $b$, $c$, $f$, and $i$—belong to Hg$^{197}$; $c$ and $f$ were observed in a weak exposure; $a$, $b$, $f$, and $i$ appeared in a strong one. Nine components should be present, but only five were observed. None of the missing components can be excluded because of low relative intensity, but $d$ and $e$ do certainly overlap the strong components, $A$ and $(196 + 198 + B)$, while $g$ and $h$ may be covered by $B$ and $f$. As a check on the five observed components we see that the difference $(i - a) = 1588$ mK agrees within 3 mK with the accepted value for the $F = 11/2 - F = 15/2$ interval in the $^3S_1$ state, however, $(b - f) = 865$ mK shows a discrepancy of 13 mK. This discrepancy may be attributed to the overlapping of $f$.

The two intervals in the $^3P_2$ state are then determined from $(c - b) = 395$ mK and from $(f + 739 - i) = 374$ mK, combined with $(b - a) = 358$ mK. Then using the $F = 13/2 - F = 15/2$ and $F = 15/2 - F = 17/2$ intervals of the $^3P_2$ state we calculate the dipole and quadrupole interaction constants for this state and obtain

$$A_{197} (^3P_2) = -49.1 \pm 0.6 \text{ mK},$$

$$B_{197} (^3P_2) = +45 \pm 12 \text{ mK}.$$

We now return to the $^3P_1$ state. From the 4358 A data we obtained $(F = 11/2 - F = 13/2) = 480$ mK and $(F = 13/2 - F = 15/2) = 606$ mK for the two intervals.

### Table I. Interaction constants for mercury isotopes.

<table>
<thead>
<tr>
<th>$^6s^7s\ ^3S_1$ (Spec.)</th>
<th>$^6s^6p\ ^3P_1$ (Spec.)</th>
<th>$^6s^6p\ ^3P_1$ (Doub. res.)</th>
<th>$^6s^6p\ ^3P_3$ (Spec.)</th>
<th>$^2S_1/^2P_1$</th>
<th>$^2P_1/^2P_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{mm}$</td>
<td>$A_{mt}$</td>
<td>$A_{nt}$</td>
<td>$A_{mt}$</td>
<td>$B_{nt}$</td>
<td>$B_{nt}$</td>
</tr>
<tr>
<td>$747.3 \pm 2$</td>
<td>$-113.6 \pm 0.5$</td>
<td>$716.93$</td>
<td>$-265.35$</td>
<td>$-26 \pm 3$</td>
<td>$-9.35$</td>
</tr>
<tr>
<td>$512.5 \pm 4$</td>
<td>$-77.8 \pm 0.4$</td>
<td>$491.87$</td>
<td>$-181.89$</td>
<td>$-111.85$</td>
<td>$-8.24 \pm 0.8$</td>
</tr>
<tr>
<td>$513.5 \pm 4$</td>
<td>$491.4 \pm 0.5$</td>
<td>$-181.7 \pm 0.5$</td>
<td>$-9.35$</td>
<td>$-8.24 \pm 0.8$</td>
<td></td>
</tr>
<tr>
<td>$-49.1 \pm 0.6$</td>
<td>$302.9$</td>
<td>$-111.85$</td>
<td>$45 \pm 12$</td>
<td>$14.02$</td>
<td></td>
</tr>
</tbody>
</table>

* The interaction constants are given in mK ($10^{-3}$ cm$^{-1}$).
The 2537 A data give 480 mK for the first and strongly support the value obtained for the second. Thus we obtain

\[ A_{197*}^{(3P_0)} = -77.8 \pm 0.4 \text{ mK}, \]

\[ B_{197*}^{(3P_0)} = -26 \pm 3 \text{ mK}. \]

With these values for the interaction constants we calculate the displacement of the \( F=11/2 \) level from the center of gravity. Then we use the quite reliable value of the \( F=11/2 \) component in the 2537 A line and obtain, for the center of gravity of \( \text{Hg}^{199} \) (with respect to \( \text{Hg}^{197*} \), \( +70 \pm 4 \text{ mK} \). For the center of gravity of \( \text{Hg}^{99} \) we use the value obtained from double resonance, which is also in agreement with the 2537 A data.

Finally, we mention a successful Zeeman exposure of the 2537 A line in a magnetic field of approximately 8 kilogauss. It was not possible to resolve the \( m \)-sublevels of \( \text{Hg}^{197*} \), which we expected to be 30-40 mK apart. Furthermore, the three \( \text{Hg}^{199} \), the three \( \text{Hg}^{201} \), and the six \( \text{Hg}^{203} \) components dominate the exposure and hide the extremely weak \( \text{Hg}^{205} \) contribution, which consists of 42 components. For these reasons, and because our experimental arrangement may have introduced field inhomogeneities of as much as 10%, we could not interpret these data.

### INTERPRETATION OF THE RESULTS

Our results are summarized in Table I, which gives the values of the dipole and quadrupole interaction constants \( A \) and \( B \) in the \( ^3S,^1P,^3P,^3P_3 \) states. We also include the interaction constants of the stable isotopes \( \text{Hg}^{199} \) and \( \text{Hg}^{201} \) as obtained spectrscopically\(^8\) and by double resonance techniques.\(^7,8\) The last two rows of the table give the ratios of the interaction constants in the three states. It is seen that agreement exists for the dipole interaction constants \( A \) and the quadrupole interaction constants \( B \).

Analysis of the data should also provide the isotope shifts for the five lines that were investigated. However, since we were not able to resolve the \( \text{Hg}^{196} \) and \( \text{Hg}^{198} \) components (except for the 2537 A line), and since the isotope shift of \( \text{Hg}^{197} \) and \( \text{Hg}^{199} \) for these lines is of the order of our experimental error, we consider the isotope shifts in the 2537 A line only. We obtained the shifts that are summarized in Table II.

Thus we observe a displacement of atomic energy levels which is due to the excitation of the nucleus from the ground to the isomeric state. The isomeric shift is \(-21 \pm 6 \text{ mK} \) for the 2537 A line. The isomeric shifts in the other lines could not be calculated accurately. They are expected to be approximately 3-4 mK.

### Table II. Comparison of isotope shifts in mercury, in mK.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Shift (mK)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{Hg}^{199} )</td>
<td>+137 ±4</td>
</tr>
<tr>
<td>( \text{Hg}^{201} )</td>
<td>+91 ±4</td>
</tr>
<tr>
<td>( \text{Hg}^{203} )</td>
<td>+70 ±4</td>
</tr>
<tr>
<td>( \text{Hg}^{205} )</td>
<td>0</td>
</tr>
</tbody>
</table>

\[ +137 \pm 4 \text{ mK} \]

\[ +91 \pm 4 \text{ mK} \]

\[ +70 \pm 4 \text{ mK} \]

\[ 0 \text{ mK} \]

The 2537 A data give 480 mK for the first and strongly support the value obtained for the second. Thus we obtain

\[ A_{197*}^{(3P_0)} = -77.8 \pm 0.4 \text{ mK}, \]

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### Table III. Quadrupole moments of \( Z=80 \) nuclei.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Spin</th>
<th>Lifetime</th>
<th>Moment (10^{-26} cm^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{Hg}^{199} )</td>
<td>3/2</td>
<td>stable</td>
<td>0.50 ±0.05</td>
</tr>
<tr>
<td>( \text{Hg}^{201} )</td>
<td>5/2</td>
<td>2.35X10^{-9} sec</td>
<td>0.71 ±0.02</td>
</tr>
<tr>
<td>( \text{Hg}^{203} )</td>
<td>13/2</td>
<td>25 hours</td>
<td>1.5 ±0.3</td>
</tr>
</tbody>
</table>

For the evaluation of the nuclear moments of \( \text{Hg}^{197*} \) we adopted the magnetic moment of \( \text{Hg}^{199} \) obtained from nuclear resonance with a diamagnetic correction\(^9\)

\[ \mu_{199} = 0.5043 \text{ nm}. \]

Then, disregarding hfs anomalies, we obtain

\[ I(197*) \times A(197*) = -1.04 \pm 0.01 \text{ nm}. \]

\[ I(199) \times A(199) \]

The magnetic moment of \( \text{Hg}^{197*} \) is closer to the Schmidt limit than is the moment of \( \text{Hg}^{201} \) (both have the same limit, \(-1.91 \text{ nm}\).) This result is to be expected because the independent-particle model should apply more accurately to a state where the optical neutron is in a high angular momentum state.

In the quadrupole moment of \( \text{Hg}^{197*} \), we followed the same procedure and adopted\(^{9} \) \( Q_{201} = (0.50 \pm 0.05) \times 10^{-24} \text{ cm}^2 \) and used the value of the interaction constant for the \( ^3P_3 \) state. Thus we have

\[ Q_{197*} = B_{197*} \times Q_{201}/B_{201} = (1.5 \pm 0.3) \times 10^{-24} \text{ cm}^2. \]

In Table III we give the three known quadrupole moments for \( Z=80 \) nuclei, two of which refer to excited nuclear states. The quadrupole moment for the \( 2.35 \times 10^{-9} \text{ sec}, I=5/2 \) state of \( \text{Hg}^{199} \) was taken from angular correlation measurements in \( \text{HgCl}_2 \) by Pound and Wertheim\(^{14}\); they calculate \( Q_{199} = \pm (0.71 \pm 0.02) \times 10^{-24} \text{ cm}^2 \).

If, contrary to the predictions of the collective model, we take \( Q_{199} \), as positive, a \( Q/R^2 \) plot (Townes plot) of the moments mentioned in Table III gives a smooth curve, which correctly decreases to zero at \( N=126 \).

With the limited data available, we do not wish to draw any conclusions about intrinsic quadrupole moments and properties of the nucleon core.

As to the isomeric shift, it represents a shift of atomic energy levels because of the excitation of the optical neutron to the \( ^3P_3 \) shell. If we assume that the excitation of the neutron leaves the nuclear charge distribution unchanged, the shift will be caused entirely by the small neutron-electron interaction. Weiner\(^{15}\) calculated the shift under that assumption and found a value smaller than 1 mK for the 2537 A line.

Thus the rather large value of \(-21 \pm 6 \text{ mK} \), which was obtained from our data, clearly indicates that the previous assumption does not hold and that the excitation


tion of the neutron is accompanied by a general redistribution of the charged nucleons. The sign of the shift indicates an increase in nuclear charge distribution that is approximately one-fourth to one-fifth of the increase resulting from the addition of one neutron.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the continuous encouragement and advice of Dean Francis Bitter, who originally proposed and started this project. We also thank Dean George R. Harrison, principal investigator of U. S. Atomic Energy Commission, for ruling the grating. We are indebted to Professor L. C. Bradley, III, for the interferometric work, and to Mr. H. Hirsch, who helped us throughout the whole project and prepared the source for one of the runs. We sincerely thank Mr. A. J. Velluto, who did all of the glassblowing for the radioactive work and whose assistance was essential to the completion of this investigation.

Note added in proof.—In reproduction for printing, some of the lines of the original spectrograms were lost; their positions are indicated by the identifying letters.