THE HIGH FREQUENCY SPECTRA OF LEAD ISOTOPES.

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Synopsis.

Least Detectable Difference in Wave-length Dependent on Distance between Lines and Length of Reflected Ray.—Reasons are given for confining the work to a comparison of the $L\alpha_1$ lines of ordinary and uranio-lead. The least detectable difference in wave-length between two lines on the same plate is directly proportional to the least detectable distance between their axes and inversely to the length of the reflected ray between the crystal and plate. However, the nature of the $L\alpha_1$ line of lead is such as to make it more important to reduce as much as possible the least detectable distance between the lines, than it is to increase the length of the reflected ray.

Limit of Accuracy. Special Method of Suspending and Displacing Plateholder.—The spectrometer is described in detail in the preceding article. A special method of suspending the plateholder and giving it a known displacement is described, by which it is possible to fix the limit of accuracy directly. An X-ray bulb of the gas type was used with a specially designed anode which could be rotated so as to bring different parts of its face under the action of the cathode stream. The two kinds of lead were placed on different parts of the same anode. The uranio-lead was from a primary uraninite from India.

Upper Half of Line from One Lead and Lower Half from Other Photographed on Same Plate.—The α_1 line of one kind of lead was photographed on the upper half of the plate, and the same line from the other lead on the lower half, immediately below it. Full lines for reference were sometimes photographed near the half lines for measurement purposes. The limit of accuracy was found by giving the plate a known displacement between the taking of one half and the other, of a line from the same kind of lead.

$L\alpha_1$ Lines from Ordinary and Uranio-lead Do Not Differ in Wave-length by as much as 0.005 Per Cent.—It is found that a displacement between two half lines of $(10)^{-3}$ mm. can be readily detected and that the wave-lengths of the $L\alpha_1$ lines of the two kinds of lead can not differ by as much as $0.6 \times (10)^{-4}$ Å., or 0.005 per cent.

Introduction.

A KNOWLEDGE of the relation between the spectrum of a substance and that of its isotope is important in that it may throw further light on the structure of the atom. Some work in this line has been done. Aronberg,[1] working with a grating spectrograph, has reached the conclusion that the wave-length of the line λ 4058 is greater by 0.0043 Å. for lead of radioactive origin than it is for ordinary lead. The work of Aronberg has recently been corroborated by Merton,[2] working with a

[1] Aronberg, Astrophysical Jour., Vol. 47, p. 96, 1918.

[2] Merton, Proc. Roy. Soc., No. A, 679, Vol. 96, p. 388, 1920.

Fabry and Perot *étalon*. The X-ray spectra of lead isotopes have been investigated by Duane and Shimizu,[1] and by Siegbahn and Stenström.[2] The former authors, using an ionization method, measured the three critical absorption wave-lengths in the L-series from a specimen of ordinary lead and one of radio-active lead, the atomic weights of which had been determined by Richards and found to differ by more than $\frac{1}{2}$ per cent. They conclude that these wave-lengths do not differ between the two kinds of lead by more than 0.1 per cent. or about 0.0008 Å. Siegbahn and Stenström used a photographic method and investigated all the lines in the L-series, and the strongest lines in the M-series of ordinary and radioactive lead. The spectra of the two kinds of lead were photographed on the same plate under the same conditions. By using a lead screen to cover one part or another of the plate at a time, the middle portion of the lines of one kind of lead were obtained with the upper and lower portions of the lines of the other kind of lead above and below. The authors conclude that the wave-lengths of the two kinds of lead do not differ by more than 0.0005 Å. They do not, however, in the article referred to, state how they estimated their limit of accuracy. The dispersion and width of slit used are not mentioned. For a description of their apparatus they refer to a number of the Jahrbuch der Radioaktivitat und Elektronik for 1916 which, up to the present, has not come to hand on account of the war.

Some time ago Professor Uhler suggested to the senior author that it would be of interest to investigate the X-ray spectra of isotopes, and as we had a supply of uranio-lead at our disposal, it was decided to undertake the work. On account of delays caused by the war we did not receive notice of the work of Siegbahn and Stenström until our work was well under way. The question of the spectra of isotopes seemed, however, of sufficient importance to warrant a repetition of the work by independent investigators even though the methods used by each were nearly the same. At the same time we had reason to believe that we could attain a sufficient degree of accuracy to detect a difference of wave-length, if present, well within the limits set by the authors named. We therefore decided to complete the work.

The potential required to produce the K-series of lead is so high that it is doubtful if this series can be produced in the ordinary gas type tube. On the other hand the wave-lengths of the L-series are long enough to make the absorption of the air an appreciable factor. Not having a vacuum spectrometer at our disposal we decided to confine our investigation to the α_1 line of the L-series of lead.

[1] Duane and Shimizu, Proc. Nat. Acad. Sci., No. 6, Vol. 5, p. 198, 1919.

[2] Siegbahn and Stenström, Phys. Zeitschr., Vol. 18, p. 547, 1917.

The $L\alpha_1$ line of lead is very broad and more resembles a band than a line. This is probably a characteristic of the lines of the L-series in general. A line of this nature will always be broader than the slit even when the plate is placed exactly at the focus of the slit, and the higher the dispersion, the greater will be this difference. The smallest difference in position between two such lines, taken in a manner similar to that described by Siegbahn and Stenström,[1] which can be detected will depend on the narrowness, sharpness, and intensity of the lines. Since these fall off as the dispersion is increased there is nothing to be gained by carrying the dispersion beyond a certain point.

If a photographic plate is so placed on the spectrometer that its plane is normal to the collimating line, and at such a distance from the axis of rotation of the spectrometer that the portion of the plate that is to receive the spectral line is either at the focus of a single slit, or somewhere between the foci of two slits of equal width, then it is easily shown that the distance, δ, between the axes of two lines whose difference in wave-length, $\Delta\lambda$, is small, is given by:

$$\delta = \frac{r\Delta\lambda}{d \cos\theta \cos 2\theta},$$

where r is the distance from the axis of rotation to the plate, measured along the reflected ray, d is the grating space of the crystal, and θ is the glancing angle. For the $L\alpha_1$ line of lead and the (100) planes of calcite, θ is approximately 11.°18 and d is 3.03 Å., which gives

$$\Delta\lambda = 2.8 \text{ Å.} \left(\frac{\delta}{r}\right). \tag{1}$$

The smallest difference in wave-length that can be detected is therefore directly proportional to the smallest detectable value of δ, and inversely proportional to r. Stated in this way however, and for the line under consideration, δ is some direct function of r, and it becomes more important to reduce δ to a minimum than to increase r.

Apparatus.

We used a spectrometer, a detailed description of which will be found in the preceding article. The plateholder was always mounted in such a manner that its plane was approximately normal to the line of collimation and at such a distance from the axis of rotation that the part of the plate where the spectral lines were produced was always either at the focus of one or other of the slits S_1 and S_2, or between these foci.

For the special purpose of this experiment we mounted a screen

[1] *Loc. cit.*

directly in front of the plateholder, but not touching it. The screen consisted of a horizontal strip of brass about 1.7 mm. thick, of about the same length, and a little more than half the breadth of the opening in the front of the plateholder. The screen was supported by uprights, which stood on the same masonry pier with the spectrometer, but which were otherwise entirely independent of the latter. It could be raised entirely above the opening in the plateholder, or lowered so as to cover either the upper or lower half of this opening as desired.

For a purpose to be explained later, the plate holder, instead of being supported on the azimuth circle clamped to the T rail, could be hung on two parallel wires of equal length, one fastened at each end of the top of the plateholder. The wires were about 134 cms. long, and their upper ends were fastened to a beam above the spectrometer. When the plateholder was supported in this way its regular mounting was removed from the T rail. In place of this the plateholder had screwed to its base a brass plate with two vertical posts extending below it and about 10 cm. apart. Through the lower end of each post and normal to the plane of the plate extended a screw with a smooth rounded point. By suitably adjusting the supporting wires, a small component of the weight of the plateholder served to make these screws bear gently against a smooth glass plate clamped to the T rail. The frame of a spherometer was clamped to one end of the T rail so that the axis of the screw was parallel to the long axis of the photographic plate, and the point of the screw bore against a smooth glass plate fastened on the end of the plate holder. The spherometer screw had a pitch of 0.5 mm. and its circular scale had a least count of 0.001 mm. To facilitate turning the screw accurately through one division of the head without rocking it in its nut, a tangent screw was provided.

For diffraction grating we used the cleavage planes of a specimen of calcite crystal which had already been found to give very satisfactory lines.

The X-ray bulbs were of the gas type and were made especially for us by Mr. A. Greiner, vice-president of the Green and Bauer Company, Hartford, Conn.; and the skillful manner in which he followed our design greatly facilitated the experimental manipulation. The one from which we obtained the fianl results had a water-cooled anode which could be rotated or removed by means of a ground glass conical joint. The anode was a copper disc with a silver disc about 2 mm. thick screwed to its face. The plane of the anode was made as nearly normal as possible to the axis of revolution of the ground glass cone. The anode was so mounted that its face made an angle of about 45° with the cathode rays, and so that different parts of its face came under the action of these rays when the ground joint was rotated. A short side tube was sealed to the main bulb so that the axis of the tube was normal to the plane containing the cathode rays and the axis of the ground joint. The end of this side tube was covered by a brass plate with a small hole in it. The hole, in turn, was covered with aluminum foil about 0.12 mm. thick. The line through this hole to the focal spot made a small angle with the plane of the anode.

The X-ray bulb was placed with the aluminium window close to the slit, S_1, of the spectrometer, with the plane of the anode vertical, and so that the line from the focal spot through the aluminium window should be as nearly horizontal as possible. It was supported on three very rigid supports clamped to the anode tube, cathode tube, and an extra side tube respectively. These supports were fastened to the same table on which the spectrometer rested. This table consisted of a heavy granite slab cemented on a brick pillar isolated from the building.

The specimens of lead to be compared were both placed on the anode at the same time in the following manner. The position of the focal spot on the anode was marked by running the bulb; the ground joint was then turned through about 45°, and the new position of the focal spot marked. The positions of the ground joint corresponding to these two positions of the anode were marked on the outer end of the cone. The anode was then removed from the bulb, and the silver disc taken off. A cluster of small holes, as close together as possible, was drilled in the silver at both positions of the focal spot. The holes were about 1 mm. in diameter, and enough were drilled to more than cover the focal spot. One of the kinds of lead to be investigated was pressed into one cluster of holes, and the other kind into the other cluster. Great care was taken not to contaminate either lead with the other.

The pressure within the X-ray bulb, while running, was maintained as nearly constant as possible by a mercury diffusion pump so that the bulb would back a 12.7 cm. parallel spark gap. The current through the bulb was about 5 milliamperes, supplied by an "Ideal Interrupterless" X-ray current generator, purchased from the Kny-Scheerer Company of New York City.

The specimen of ordinary lead was cut from ordinary commercial sheet lead taken from the stock in the laboratory. The uranio-lead was very kindly prepared and furnished by Professor B. B. Boltwood with the statement that it was obtained from some lead chloride that had been separated from a primary uraninite from India; that it should be entirely free from ordinary lead, and should contain only a little "thorio-lead," if such a substance exists.

Method of Comparing Spectra.

The method employed to compare the spectra of the two leads was as follows. The plateholder was placed on its usual mounting on the T rail, and one half of its breadth covered by the brass screen. It was then exposed to the rays from one kind of lead, the crystal always being given a small rotation back and forth around the position which gave the maximum reflection. At the end of the exposure the anti-cathode was turned until the other kind of lead came under the focal spot, and the plate exposed again with its other half covered by the screen. On some plates a full line was taken on each side of the half lines and about 2 mm. from them. This was to give fiducial lines from which to measure. Plates were taken at various distances from the axis of rotation of the spectrometer and with various distances between this axis and the slit S_1. The distance from the axis to slit S_2 was always 4.5 cms. Different widths of slit were tried, and sometimes one, and other times two slits were used.

The plane of the anode did not remain absolutely fixed when the ground joint was turned to bring one or the other lead under the focal spot, though the variation was very slight. However, when using two very narrow slits a long distance apart, it was necessary to realign the spectrometer after turning the anode. This could easily be done by rotating the base of the spectrometer about the long axis of S_1 as described in the previous article. The spectrometer was lined up for one kind of lead and the position of the base noted. The other kind of lead was then brought into position, and the spectrometer realign for it. The first half line was then taken without changing the anode. Then, after the first kind of lead had been brought back into position, an auxiliary plate was introduced close to S_2 and a series of exposures of equal length taken on it for different recorded positions of the spectrometer base around the position which had been noted. The position which gave the most intense image of S_2 was then used in taking the second half ine. Absolutely no part of the spectrometer, slits, crystal or plateholder, were ever touched from the time the exposure for the first half line was started till the exposure for the second half was completed, except to rotate the base of the spectrometer to adjust the line up. This was done by slow motion screws, the base resting at two points on *plane* surfaces and pivoted at a third point on a carefully made cone. The auxiliary plate was supported without touching any part of the spectrometer.

Test for Limit of Accuracy.

We wished, if possible, to compare the spectra of the two kinds of lead to a much higher degree of precision than had previously been attained. We were therefore confronted with the problem of finding what was the smallest displacement that we could detect between two half lines taken in the manner described. To do this we proposed to give the photographic plate a known displacement after taking one half line and before taking the other, both halves being produced by the same kind of lead. Various means of supporting and displacing the plateholder were tried for this purpose and proved unsatisfactory. Finally, at the suggestion of Professor Uhler, we hung the plateholder on wires in the manner already described and displaced it with the spherometer screw.

Results.

1. *Limit of Accuracy.*—Two plates, nos. 41 and 44, were taken with the plateholder suspended on the wires. Slit S_1 was wide open, and S_2 was 0.02 mm. wide. The value of r, equation (1), was 4.5 cms. The crystal was rotated back and forth through an angle of $1\frac{1}{2}'$ on each side of the best setting. On No. 41 five exposures were taken using the uranio-lead only. The first exposure was a full line, the third and fourth were upper halves, and the second and fifth were lower halves. The plateholder was displaced by the screw in the same direction after each exposure; 2.000 mm. after the first, 0.002 mm. after the second, 2.000 mm. after the third, and 0.004 mm. after the fourth. On No. 44 only four exposures were taken, using the ordinary lead. The first and fourth were full lines, the second was a lower, and the third an upper half. The displacements after the first and third were both 2.000 mm., and that after the second was 0.001 mm.

Both the small displacements between adjacent half lines on plate No. 41 were quite obvious under a low power magnifying glass. The displacement of 0.001 mm. on No. 42 was not visible in this manner. The lines on both plates were sharp and black and not more than 0.025 mm. broad. Both plates were measured on a measuring engine, frequently tested and regularly used by Uhler and others for measuring plates obtained with the large Rowland spectrograph. The measurements were made along two lines normal to the spectral lines, one a small distance above the break between the halves and the other the same distance below. The measured values of the distances agreed very closely with the actual displacements. The values of the small displacements, obtained by subtracting the upper from the lower distances, in no case differed from the actual displacements by as much as 0.001 mm. We therefore feel justified in assuming that with lines of the sort obtained on these two plates, a displacement of 0.001 mm. could not escape detection.

2. *Comparison of Leads.*—The following plates were taken with two exposures, giving the upper half of the line from one lead and the lower half from the other, on each, the plateholder remaining fixed during the two exposures, and the crystal rotated back and forth around the best setting.

Plate No. 6.—The width of S_1 was 0.04 mm., and S_2 was wide open. The distance from S_1 to the axis of rotation was 35 cms. and the value of r was 35 cms. The ordinary lead line was above, and the uranio-lead line below. The lines were very fuzzy, though dark, and about 0.1 mm. broad.

Plate No. 7.—The data for this plate are the same as for No. 6 except that S_1 was closed to 0.02 mm. and the positions of the lines from the two leads were reversed. The lines were not quite as broad as on plate No. 6, but were weaker and just as fuzzy.

Plates No. 29 *and No.* 30.—The width of both S_1 and S_2 was 0.02 mm. The distance of S_1 to the axis of rotation was 20 cms. and the value of r was 20 cms. The positions of the lines were reversed on the two plates. The lines were narrower than on the other two plates and were fairly strong, but were still too broad and fuzzy to permit of precise measurements.

None of these plates showed any displacement between the lines, but their axes were too indefinite to permit of their being compared with a high degree of precision. An attempt was made to set a superior limit to the displacement that could exist without being visible. These plates were placed on the measuring engine, and one cross hair of the microscope was set as nearly as possible parallel to, and on what appeared to be the axis of the pair of half lines considered as one continuous line. The engine screw was then turned the least amount that would throw the cross hair, beyond question, off the axis of the lines. In this manner it was estimated that, on the first two plates, there could not have been a displacement as great as 0.02 mm., and on the second two, as great as 0.01 mm. If these values are assigned to δ in equation (1), with the corresponding values of r, it is seen that the possible difference of wave-length between the lines of the two kinds of lead can not exceed $1.6\ (10)^{-4}$ Å. on the first two plates, and one half this value on the second two.

In order to have lines which could be measured with as high a degree of accuracy as those obtained in the tests for the smallest detectable displacement, we took two more plates, No. 46 and 47. The conditions under which these plates were taken were as follows. Both slits were 0.02 mm. wide. The distances from S_1 and S_2 to the axis of rotation were 15.6 cms. and 4.5 cms. respectively. The value of r was 4.9 cms. The crystal was rotated back and forth through an angle of $1\frac{1}{2}'$ on each side of the best setting. Four exposures were taken on each plate, namely: a full line A was taken for one kind of lead, the plateholder was displaced along the T rail about 2 mm., and the lower half, B, of the line from the same kind of lead was taken; the plate holder remaining fixed, the upper half, C, of the line from the other kind of lead was taken, and then the plateholder was displaced about 2 mm. further and a full line, D, from the same lead as C, was taken. The direction of the displacement of the plateholder was such as to place A on the long wave-length side of B and C, and D on the short.

Table I.

Plate No. 46.

Upper Half.			Lower Half.		
(AC).	(AD).	(CD).	(AB).	(AD).	(BD).
2.1355	4.2021	2.0666	2.1384	4.2038	2.0654
.1341	.2033	.0692	.1373	.2051	.0678
.1370	.2038	.0668	.1356	.2035	.0679
.1358	.2059	.0701	.1363	.2061	.0698
. .1363	.2043	.0680	.1357	.2048	.0691
Mean 2.1357	4.2039	2.0681	2.1367	4.2047	2.0680

$(AB) - (AC) = 0.0010$
$(CD) - (BD) = 0.0001$
Mean $= 0.0006$

Plate No. 47.

Upper Half.			Lower Half.		
(AC).	(AD).	(CD).	(AB).	(AD).	(BD).
2.0020	4.0512	2.0492	2.0045	4.0534	2.0489
.0045	.0522	.0477	.0075	.0550	.0475
.0044	.0522	.0478	.0021	.0538	.0517
.0064	.0550	.0486	.0046	.0539	.0493
.0036	.0524	.0488	.0057	.0533	.0476
.0031	0.517	.0486			
Mean 2.0040	4.0525	2.0485	2.0049	4.0539	2.0490

$(AB) - (AC) = 0.0009$
$(CD) - (BD) = -0.0005$
Mean $= 0.0002$

On plate No. 46 the lines A and B were produced by uranio-lead and the lines C and D by ordinary lead. On plate No. 47, on the other hand, the lines A and B were produced by ordinary lead, and C and D by uranio-lead. The lines on both plates were well defined and sharp though not quite as black as on plates 41 and 44. The upper and lower halves of each plate were measured on the engine. These measurements are set forth in detail in Table I., the figures in each column being the various measurements of the distance between the two lines corresponding to the letters at the head of the column. The units are approximately millimeters.

It will be seen from the table that on plate No. 46 the measurements from both A and D assign to C, an ordinary lead line, a longer wave-length than B, a uranio-lead line. The measurements from A on plate No. 47, however, assign to C, a uranio-lead line, a longer wave-length than B, an ordinary lead line, while the measurements from D reverse this order. None of the displacements between the half lines deduced from the measurements of these plates are greater than 0.001 mm., and the means are much less. Obviously the conclusion to be drawn from these two plates is that there is no difference in wave-length between the two kinds of lead great enough to produce a shift in the line of as much as 0.001 mm. With the value of r used with these plates we feel justified in the conclusion that the difference in wave-length between ordinary and uranio lead for the α_1 line of the L-series can not be as great as 0.00006 Å. The wave-length of this line being about 1.18 Å., the difference, if it exists at all, must be less than 0.005 per cent.

In conclusion it gives us great pleasure to express our sense of obligation to Professor Uhler for his interest and many valuable suggestions in connection with the work, and to Professor Boltwood for supplying us with the specimen of uranio-lead used.

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