Very Soft X-Ray Spectra of Heavy Elements

By C. DEL ROSARIO University of Pennsylvania

(Received May 31, 1932)

Absolute wave-length measurements by photographic methods with plane gratings (280 and 1180 lines per mm) have been made on very soft x-ray lines of Th, Pb, Hg, Au, Ir, W and Ta. The wave-lengths range from 43.6A to 104.8A. For Hg, Au, Ir, W and Ta doublets were observed whose probable origins are transitions within the N shell, i.e., $\Delta n = 0$. These doublets fall on a smooth Moseley curve of small curvature.

INTRODUCTION

IN A recent work¹ on very soft x-rays, Thibaud measured the wave-lengths of several emission lines of the N and O series of some elements of high atomic numbers. He showed that, from the ordinary x-ray selection rules and with the energy-level values computed by Bohr and Coster, no electron transition could be selected that could predict accurately the wave-lengths of several lines he obtained by experiment. He suggested the possibility of an electron transition from one orbit to another in the same shell, and he showed a good agreement between the energy changes following such *forbidden* transitions and those experimentally observed. On account of the fundamental character of the principles and facts involved, it was thought desirable to confirm and extend Thibaud's determinations with hope of obtaining additional information on the structure of the outer part of the heavy atoms.

EXPERIMENTAL METHODS

The high absorbability of the radiation to be investigated requires the experiments to be done in vacuo. In the present work photographs of the



x-ray lines were taken with the aid of two vacuum spectrographs of a design resembling in general principles those used by Thibaud² and other investigators³ in soft x-rays. Several variations in details were made, however, with the end in view of possible improvements or simplifications. Fig. 1 shows

¹ Thibaud, Comptes Rendus 188, 1394 (1929).

² Thibaud, Jour. de Physique 8, 447 (1927).

³ For instance see: Hunt, Phys. Rev. **30**, 227 (1927); Weatherby, Phys. Rev. **32**, 707 (1928); Howe, Phys. Rev. **35**, 717 (1930); Soderman, Zeits. f. Physik **65**, 656 (1930).

137

schematically the principle involved in the wave-length measurements. X-rays from the target T pass through the limiting slits S_1 and S_2 . Part of the narrow beam thus formed falls on a glass line grating G which reflects and diffracts it, while the rest passes by the grating and falls on a photographic plate P at A. The reflected and diffracted beams fall on the plate at B and C respectively.

The wave-lengths of the lines were calculated from the ordinary diffraction relation

K

$$b\lambda = D \left[\cos \theta - \cos \left(\theta + \alpha \right) \right]$$

using the modified form $n\lambda = 2D \sin \left[(2\theta + \alpha)/2 \right] \sin \alpha/2$ where *n* denotes the order of the spectrum, λ , the wave-length and *D* the grating constant. To determine the glancing angle θ , it is necessary to know the position on the photographic plate of that part of the x-ray beam which was reflected and diffracted. This was accomplished by giving to the position of the image of the undeviated beam such correction as to make the wave-lengths of the different orders of a line—e.g., the O $K\alpha$ —agree among themselves. This correction was always small. Most of the wave-length measurements were made with the nearest order of this line for comparison.

The glass gratings used in this work have spacing constants of 4.000×10^{-4} cm and 8.476×10^{-5} cm determined with the aid of the 5461A line of mercury. The grating was mounted on a carefully made holder which, by fine screws, can give it rotation about three mutually perpendicular axes besides a horizontal translation perpendicular to the x-ray beam without disturbing the orientation of the grating.

The limiting slits have beveled jaws with fine screws for width adjustment and horizontal displacement perpendicular to the x-ray beam. In addition, both slits can be rotated independently about an axis parallel to the x-ray beam to put them in the same vertical plane.

The photographic plate is held perpendicular to the x-ray beam in a lighttight box with the emulsion side pressed against a rigid metal surface in the box, obviating the necessity of measuring the thickness of the glass backing of every plate to get the grating-to-plate distance. The box is provided with a trap door in the front end which can be opened and closed at will by a magnet outside the spectrograph. One of the boxes used was so constructed that two parallel plates at different distances from the grating could be exposed at the same time. This offered a way to check up the measured distance, grating to plate, the distance between the plates being known. Five inch plates were used in the big spectrograph at about 50 cm from the grating, and lines as far as 10 cm from the undeviated line could be measured. The positions of these lines were obtained from densitometer measurements.

Fogging of the plates due to stray visible light from the hot filament, which gave trouble to some previous investigators, was effectively eliminated by three circular disks. They were bolted to the optical bench, which also carried the slits and grating holder. Two of these disks, each with an opening 1.5×0.2 cm, were placed one in front of each slit; while the third, which had a rectangular opening, part of which was covered with aluminum foils of suitable thickness to cut down the intensities of the undeviated beam and the

C. del ROSARIO

reflected beam, was in front of the plate box. The undesirable broadening of the images of the direct and reflected beams was prevented by the aluminum foils.

Figs. 2 and 3 show the construction of the two types of x-ray tubes employed. Copper, iron, lead, bismuth, iridium and gold targets were used, the last four in the form of buttons imbedded in copper. The electron source was a spiral filament of pure tungsten, thoriated tungsten or tantalum. It can



be seen that the target can easily be removed from the tube for cleaning or replacement, and put back in its original position. The filament can also be replaced readily. Its position and that of a focussing tube were adjusted by a series of trials to make the focal spot of the most satisfactory size. The wide flange at the outer end of the glass insulator for the target in the metal x-ray tube increases greatly the sparking distance, while the turned-in edges at the inside end keep the inner surface of the insulator free from sputtered metal.



The vacuum in the spectrograph and in the x-ray tube was obtained with a Kurth two-stage mercury vapor pump backed by a Cenco Megapump. Voltages ranging from 2500 to 7500 volts were supplied to the x-ray tube by a transformer-rectifier-condenser set connected for full wave rectification. The current through the tube ranged from 30×10^{-3} to 100×10^{-3} ampere, but in no case was the power used more than 300 watts.

EXPERIMENTAL RESULTS

Table I gives the wave-lengths of the observed lines, the corresponding values for ν/R and the probable transitions giving rise to these lines. For comparison, Thibaud's wave-lengths are also listed as well as computed values of ν/R based on Bohr and Coster's table of energy levels.

DISCUSSION OF RESULTS

The agreement between the wave-lengths here obtained and those of Thibaud for the same lines is very satisfactory. The two observed lines each of tantalum, tungsten, gold and mercury seem to form regular or relativity doublets of about 3A separation. The transitions assigned to these lines are identical for these elements but are forbidden by the selection rules, since

Wave-length					
Element atomic number	Present work	Thibaud's value	Probable origin	Present work	Bohr and Coster
Tantalum 73	58.3A 61.3	58.3A 61.4	$rac{N_{iv} - N_{vi}}{N_v - N_{vi-vii}}$	$\begin{array}{c} 15.6\\ 14.8\end{array}$	$15.6\\14.4$
Tungsten 74	56.0 59.0	56.0 59.1	$\begin{array}{c} N_{iv} - N_{vi} \\ N_v - N_{vi-vii} \end{array}$	$\begin{array}{c} 16.2 \\ 15.4 \end{array}$	$\begin{array}{c} 15.9 \\ 15.6 \end{array}$
Iridium 77	53.0		$N_v - N_{vi-vii}$	17.2	
Gold 79	$\begin{array}{c} 46.2\\ 48.9 \end{array}$	$\begin{array}{c} 46.8\\ 49.4\end{array}$	$\begin{array}{c} N_{iv} - N_{vi} \\ N_v & - N_{vi-vii} \end{array}$	$19.7\\18.6$	$\begin{array}{c} 20.0 \\ 18.6 \end{array}$
Mercury 80	$\begin{array}{c} 43.6\\ 46.4\end{array}$		$\begin{array}{c} N_{iv} - N_{vi} \\ N_v & - N_{vi-vii} \end{array}$	$\begin{array}{c} 20.8 \\ 19.6 \end{array}$	-
Lead 82	$84.2 \\ 102.3 \\ 104.8$		$\begin{array}{c} N_{vi} - O_{iv} \\ O_i - P_{ii} \\ O_i - P_{iii} \end{array}$	$10.8 \\ 8.9 \\ 8.7$	10.0
Thorium 90	$50.4\\64.5\\68.3$	$50.3 \\ 64.5 \\ 68.1$	$\begin{array}{c} N_{vii} - O_{v} \\ O_{ii} - P_{i} \\ O_{iii} - P_{i} \end{array}$	18.1 14.1 13.3	18.4

TABLE I. Wave-length of observed lines.



here the total quantum number does not change. The nearest values for changes in ν/R allowed by the rules are 1.9 to 2.6 or 9.5 to 17.6 percent smaller than those corresponding to the observed lines which, by the way, are the only ones found for these elements. Fig. 4 shows how closely these doublets follow Moseley's law.

No spectrum line has so far been observed characteristic of bismuth. This is not surprising since its low melting point and poor heat conductivity made it impossible to use a fair amount of power in the x-ray tube and to use a reasonably small focal spot. The lines obtained for iridium, mercury and lead C. del ROSARIO

are believed to have been measured for the first time. There is some doubt about the justification of assigning to mercury the lines at 43.6A and 46.4A. The assignment was made on the basis of known facts for the doublets of tantalum, tungsten and gold. First, the two lines have about the same separation as the other doublets; second, the component of greater wave-length is of greater intensity; third, the plotted points fall close to the Moseley curve for the other elements if the atomic number for mercury is used. The line at 53.0A is probably the more intense line of the doublet for iridium. Why corresponding doublets did not appear in the spectra of lead and thorium is not yet understood; however, a further extension of this work may shed light on this question.

In conclusion I wish to express my thanks to Professor C. B. Bazzoni for his encouragement and suggestions during the progress of the work.

140