

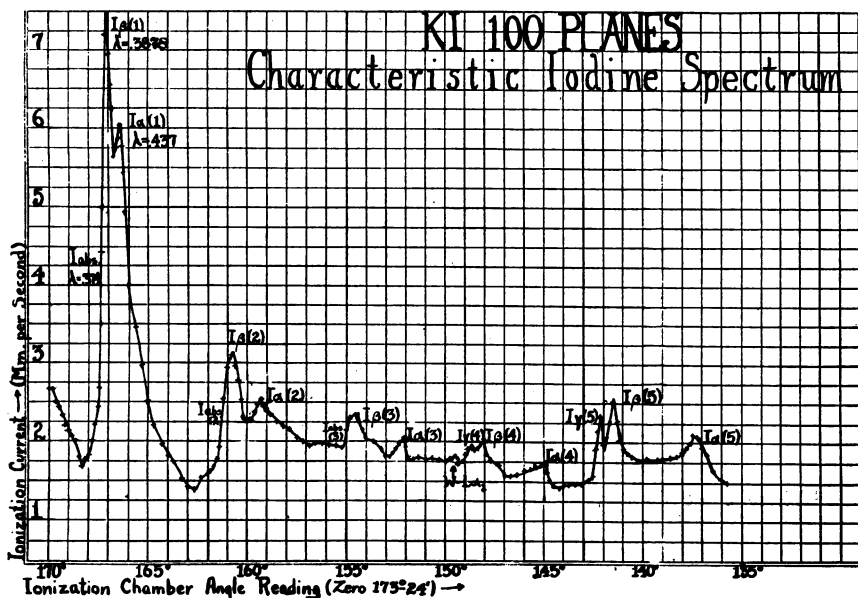
FURTHER EXPERIMENTS UPON THE REFLECTION BY A
CRYSTAL OF ITS CHARACTERISTIC X-RADIATION

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Communicated, December 5, 1923

During the development of a new method of analyzing crystals by X-rays we discovered that the X-rays characteristic of and excited in the chemical elements in a crystal are reflected by the crystal itself. The reflection takes place both in accordance with the Bragg law: $n\lambda = 2d \sin \theta$ and also in an anomalous fashion. The ionization spectrometer utilized in these investigations and the various steps in the method of analyzing crystals have already been described in detail.¹ From the spectra of the normally reflected X-rays characteristic of the constituent elements in the crystal we have been able to determine accurately the spatial distribution of the atoms in the crystal lattices of potassium iodide, cesium iodide, potassium tri-iodide, cesium tri-iodide and cesium dibromo-iodide, and to distinguish clearly between planes containing iodine and cesium atoms in spite of the nearly identical reflecting powers.



It is the purpose of the present note to present (1) a new precision measurement of the spectrum, characteristic of iodine, reflected through 5 orders

by the 100 planes of potassium iodide in accordance with the Bragg law, and (2) a further verification of the *anomalously* reflected X-rays characteristic of iodine from potassium iodide.

A crystal of pure potassium iodide, 2 mm. square and .75 mm. thick, was set up on the crystal table of the ionization spectrometer. A narrow beam of the general radiation from a tungsten target struck the crystal in such a way as to be reflected from the 100 planes into the ionization chamber. The spectrum of this reflection appears in figure 1. The mirror image of this curve has been obtained on the opposite side of the zero or direct beam zone. The angular deviations from the zero of the ionization chamber (at $173^{\circ}24'$) give the values of 2θ , since the abscissas refer to ionization chamber angles.

The following table presents the essential data involved in the spectrum curve in columns as follows: (1) ionization chamber angle reading; (2) the angle of incidence, θ ; (3) the wave-length, λ ; (4) the kind of radiation; (5) the order of the spectrum; and (6) the calculated value of θ , taking d_{100} for potassium iodide as 3.532 ångströms.

DATA RELATIVE TO THE REFLECTED IODINE RADIATION					
IONIZATION CH. ANGLE	ANGLE, θ , OBSERVED	RECORDED WAVE-LENGTH	KIND OF RADIATION	ORDER OF SPECTRUM	ANGLE, θ , CALCULATED
167°20'	3° 2'	.3737	I abs.	1	3° 2'
167° 6'	3° 9'	.3878	I β	1	3° 8'50"
166°18'	3°33'	.437	I α	1	3°33'
161°15'	6° 4' ¹ / ₂	.3737	I abs.	2	6° 4'30"
160°47'	6°18' ¹ / ₂	.3878	I β	2	6°18' 5"
159°12'	7° 6'	.437	I α	2	7°6' 25"
155° 5'	9° 9' ¹ / ₂	.3737	I abs.	3	9° 8'
154°27' ¹ / ₂	9°28' ¹ / ₄	.3878	I β	3	9°28'45"
152° 0'	10°42'	.437	I α	3	10°41'40"
149°20'	12° 2'	1.473	W L α_1	1	12° 2'
149° 0'	12°12'	.3737	I abs.	4	12°13'
148°30'	12°27'	.3818	I γ	4	12°29'
148° 0'	12°42'	.3878	I β	4	12°40'40"
144°50'	14°17'	.437	I α	4	14°19'30"
142° 5'	15°39' ¹ / ₂	.3818	I γ	5	15°40'
141°30'	15°57'	.3878	I β	5	15°55'30"
137°20'	18° 2'	.437	I α	5	18° 1'

Zero of ionization chamber = $173^{\circ}24'$.

Wave-lengths are expressed in ångströms.

Grating space $d = 3.532$ ångströms.

The experimental results presented in figure 1 and the table lead to the following interpretations: five groups of peaks appear, marked by sharp absorption drops on their short wave-length sides. The wave-length corresponding to each discontinuity has been absolutely determined by setting on each point, lowering the voltage applied to the X-ray tube in steps until the ionization current disappeared,

and calculating from the quantum equation, $Ve = h\nu$, or $\lambda = hc/Ve$. Representative critical voltages for particular points in figure 1 are 33,030 (abs. drop at $167^{\circ}20'$), 32,300 (peak at $142^{\circ}5'$), 31,800 (peak at $141^{\circ}30'$) and 28,250 (peak at $137^{\circ}20'$), respectively. These voltages correspond to the wave-lengths .3737 (I abs.), .3818 ($I\gamma$, detected and measured for the first time), .3878 ($I\beta$) and .437 ($I\alpha$), all in the K series. The spectrum represents, therefore, the characteristic K series of the iodine atoms in the crystal of potassium iodide. Substituting the wave-lengths and corresponding angles in the Bragg equation, $n\lambda = 2d \sin \theta$, gives for the average value of d , 3.532 ångströms, a value identical with that previously obtained with a wave-length from the continuous spectrum. The reflection of these characteristic rays is, therefore, regular in the sense that they obey the Bragg law. On comparing columns (2) and (6) in the table it appears that any particular point will give a value of d within a small fraction of 1% of 3.532, since the agreement between θ observed and θ calculated from this value of d is very satisfactory. The slight discrepancies are not systematic indicating that the values of λ and of d are as nearly correct as may be ascertained from such experiments.

The spectrum appears only when the X-ray tube is operated above 33,000 volts; i.e., the primary beam must contain wave-lengths equal to or shorter than the critical absorption wave-length of iodine. We used 65,000 volts in obtaining the spectrum shown here. In other experiments with voltages above 69,000 volts, the tungsten line spectrum also appeared mixed with the iodine spectrum. Even at 80,000 volts, however, the latter is much more intense than the tungsten spectrum. A comparative energy study of the primary beam at 50,000 and 78,400 volts shows an enormous difference in the region of the spectrum shorter than the critical absorption wave-length of iodine, .3737 ångström—a fact which is of great importance in the production and detection of both anomalously and regularly reflected iodine rays. Curves illustrating these comparisons will be published soon.

Faint evidence of the strong $L\alpha_1$ line of tungsten at $149^{\circ}20'$ exists. The great intensity of the iodine spectrum and the enormous absorption of the crystal for long waves, however, practically blot out the tungsten L spectrum.

The crystal of potassium iodide may be said to be fairly opaque to rays shorter than .3737 ångström, i.e., to those reflected at angles smaller than $3^{\circ}2'$. In experiments with voltages above 69,000 volts the first order tungsten K -lines at about half this angle are of feeble intensity. This illustrates the possibility of serious errors in crystal structure analyses where relative intensities of spectral lines are involved, if these lines happen to have wave-lengths in the neighborhood of the critical absorptions of the elements in the crystal. Mie,² for example, has recently corrected such

errors by a study of these characteristic spectra, excited and reflected by crystals, that we discovered some years ago.

The characteristic iodine spectrum lines are more diffuse than the regularly reflected lines characteristic of the X-ray tube target. We have never been able, by the most careful measurements, to completely separate α , β and γ peaks in the first order. The α and β peaks are practically separated in the second order, and β and γ are separated partially in the fourth order and completely in the fifth order. The α peak is not resolved into α_1 and α_2 even in the fifth order.

The relative intensities of the various characteristic lines are quite different from those in all target and secondary fluorescent spectra. The β peak is always higher than the α , while for spectra characteristic of targets, the intensities stand in the order: $\gamma:\beta:\alpha_2:\alpha_1 = 15:35:50:100$.³

I— $K\gamma$ with a wave-length .3818 ångstrom is isolated and measured for the first time. It has also been identified, with the same wave-length, in the secondary fluorescent spectrum of iodine from potassium iodide due to primary radiation from a tungsten target, the secondary radiation being analyzed by a calcite crystal spectrometer. (These PROCEEDINGS, Dec. 1923.)

The relatively great intensity of the fifth order peaks is undoubtedly connected with the fact that within this angular range the anomalous reflection of iodine rays seems to coincide with the regular reflection of iodine rays from the 100 planes. A diagram of the relative positions of

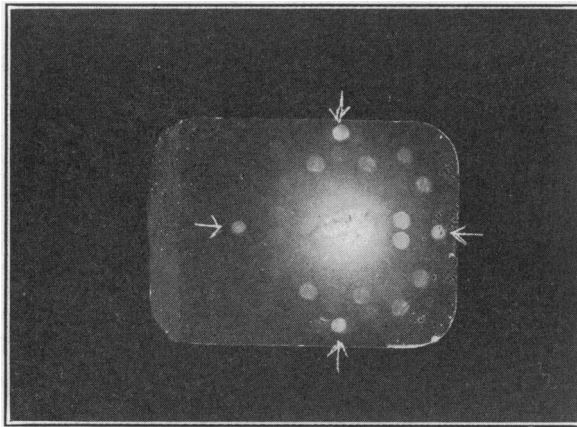


FIGURE 2

the so-called X-peaks as they depend on the angle of incidence has been published in these PROCEEDINGS, April 1923, page 133. Here the extrapolated X-peak lines intersect the line representing reflection from the

100 planes at an angle (ionization chamber) of about 32° . Table I shows that $I\beta$ (5) appears at an equal angle, 2θ , of $31^\circ 54'$.

Although one experimenter has not been able to detect this anomalously reflected characteristic radiation⁴ a repetition of our experiments with the ionization spectrometer and with Laue photographs has verified completely our previous results.⁵ A Laue photograph of the potassium iodide crystal described above is reproduced here (Fig. 2). The essential experimental details are as follows: one pin-hole, 1.6 mm. in diameter in a lead plate directly in front of crystal; film 3 cm. behind crystal; beam parallel to a principal axis; tungsten-target tube operated at a non-fluctuating potential difference of 78,400 volts for 8 hours; target crystal distance = 75 cm. and tube so placed that X-rays passing through pin-hole almost grazed the target's surface on leaving it. Some 60 spots appear on the photograph itself in addition to the central image of the direct beam. Most of these probably are due to regular reflection of particular wavelengths in the general radiation by various sets of parallel planes. Four spots (indicated by arrows), which are not less intense than any others, appear above, below, right and left, on lines bisecting the photograph at right angles, each at a distance of about 1.3 cm. from the center. The angle, whose tangent is one-third of 1.3 = .4333, is about $23^\circ 30'$, which agrees closely with the value of $23^\circ 36'$ obtained from the diagram mentioned above. Thus, when the 100 planes are at $\theta = 0$, both the ionization chamber and the photograph register the anomalously reflected iodine rays at about $23^\circ 33'$.

That the reflection is not an exact phenomenon is proven by the fact that when two pin-holes define the incident beam upon the crystal instead of one, the Laue spots of X-peaks are so greatly cut down in intensity as compared with the regular reflections that they are barely visible after 8 hours' exposure. As pointed out above, the reflection of rays characteristic of elements in a crystal by the same crystal is not exact, but somewhat diffuse.

There are some features of the Laue photographs which are difficult to explain. Without definite experimental knowledge of the wave-lengths producing given spots, such as is possible in our ionization spectrometer method, caution must be exercised in interpreting Laue photographs as a method of studying crystal structures. Wave-lengths have sometimes been assigned to spots which are actually not present in the primary beam because of an operating voltage too low to produce those wave-lengths at all. Furthermore, it is often necessary to use a voltage considerably above a critical value in order to give sufficient intensity for photographic analysis. Thus, though the critical voltage for the X-spots is 33,000, or that required to produce the critical absorption wave-length of iodine (.3737 ångström), 75,000 volts produces a beam very much richer in rays with

this and shorter wave-lengths and, consequently, the effect is much greater. Since our discovery of the anomalously reflected iodine rays from potassium iodide, several cases of anomalous reflection have been reported and explanations suggested by McKeehan⁶ on several metals, Dickinson⁷ on potassium iodide and tin tetra-iodide, and Mie² on bismuth, etc. Apparently, the explanations are not adequate, however, for the particular cases discussed here.

The excitation of X-rays characteristic of constituent elements and their regular reflection by the same crystal seems to be reasonably well explained on the basis of the principle of the transfer of radiation momenta in quanta.⁸ This analysis alone accounts for the facts, including the peculiar relative intensities, the diffuse nature of the lines, etc. Mie² considers the phenomenon as true optical resonance. If this is true, it should be possible, by using a voltage between 28,250 and 31,800, to produce an α peak without a β or γ peak. There is every reason to believe from our previous researches that the α line cannot be produced without the β and γ , just as it cannot be produced alone by the target. Furthermore, if spherical characteristic wave-trains spreading out from atoms in the crystal and reflected from other atoms produced these effects, then they ought not to depend much upon the angle of incidence of the primary X-ray, which they do, according to our experiments.

¹ Clark and Duane, These PROCEEDINGS, 8, 90 (1922); 9, 117, 126, 131 (1923); *J. Optical Soc. Amer.*, 7, 455 (1923); *Science*, 58, 398 (Nov. 13, 1923).

² *Zeit. Physik*, 15, 56; 18, 105 (1923).

³ Duane and Stenström, these PROCEEDINGS, 6, 477 (1920).

⁴ Wyckoff, *Science*, July 20, 1923.

⁵ Clark and Duane, *Science*, Nov. 13, 1923.

⁶ McKeehan, *J. Optical Soc. Amer.*, 6, 989 (1922).

⁷ Dickinson, *Phys. Rev.*, Aug. 1923, p. 199.

⁸ Duane, these PROCEEDINGS, 9, 158 (1923).

THE LAW OF SUN-SPOT POLARITY

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Communicated, December 6, 1923

In a paper on "The Direction of Rotation of Sun-spot Vortices," published in the *Proceedings of the National Academy of Sciences* in June, 1915, I described the general reversal of the magnetic polarity of sun-spots observed at the minimum of solar activity in 1912. After the usual interval of about eleven years another minimum has occurred, and the incoming