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ENERGY OF $K\alpha_3$ OF COPPER AS A FUNCTION OF APPLIED VOLTAGE WITH THE DOUBLE CRYSTAL SPECTROMETER

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Abstract

With a double crystal spectrometer of special design here briefly described the following questions were investigated with a view to determining the origin of the satellite $K\alpha_3$ of copper.

(1) Dependence of intensity of satellite $K\alpha_3$ of copper on voltage with constant current.

(2) Dependence of intensity of parent line $K\alpha_1$ of copper on voltage with constant current.

(3) Ratio of intensities of satellite to parent.

(4) Dependence of satellite intensity on current at constant voltage.

The conclusions are (1) that the satellite is excited at a voltage differing from the excitation voltage of the parent line by too small an amount to be measured with certainty (less than 200 volts). (2) That the ratio of intensities of α_1 to α_3 as estimated from the *areas* of the spectral line structures is about 1:120. (3) That the intensity of the satellite $K\alpha_3$ of copper is strictly proportional to the current at constant voltage.

These facts seem to invalidate the Wentzel-Druyvesteyn "spark line" hypothesis as an explanation of $K\alpha_3$ of copper. Richtmyer's "double jump" hypothesis remains tenable.

In addition a doublet structure (separation of components about 2 X.U.) in $K\alpha_3$ of copper was observed (in accord with the doublet structure of this satellite in elements of lower atomic number).

Introduction

THE work described in this article was undertaken with the idea of testing whether the Wentzel-Druyvesteyn theory of x-ray satellites can explain the origin of the satellite $K\alpha_3$ of copper. Richtmyer[1] has recently proposed an alternative theory of the origin of satellites and the authors of this paper are indebted to his able discussion of the question for the stimulus to undertake this research.

According to the Wentzel-Druyvesteyn "spark line" theory the satellites are to be ascribed to electron transitions between multiply ionized states. The difference in frequency between the satellite and parent line is thus ascribed to a change in nuclear screening. This would require the atom emitting such a satellite to have lost at least two electrons from the high energy states (inner levels). As Richtmyer points out, this can occur either in two successive processes in which case the satellite should be extremely faint relative to the parent line and should vary in intensity as the square of the x-ray tube current or in a single process in which case the excitation potential of the satellite must be considerably higher than that of the parent line.

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[1] Richtmyer, Journal Frank. Inst. **208**, 325–361 (1929).

On Richtmyer's "double-jump" theory of satellites on the other hand the atom emitting the satellite is supposed to execute two electron transitions which simultaneously cooperate to produce one quantum of radiation. One of these transitions has as final level the level belonging to the series of the parent line. The other transition is one of relatively small energy between two outer atomic levels. Assuming that an atom is prepared for such a state of affairs by ionization of an outer and an inner level *in one process*, only very small differences need exist between the excitation potentials of satellites and their parent lines.

In view of these considerations, we have therefore in this research investigated the following questions.

(1) Dependence of satellite intensity on voltage with constant current.
(2) Dependence of parent line intensity on voltage with constant current.
(3) Ratio of intensities of satellite to parent line.
(4) Dependence of satellite intensity on current at constant voltage.

Description of Apparatus

The complete double crystal spectrometer outfit used in this work is of a new and original design and as a detailed description of it is to be given in a separate article soon to be published, it will only be briefly described here.

The ionization chamber is solidly fixed on top of the Hoffman vacuum binant electrometer with a very short wire connecting the collector to the suspension of the electrometer through a large diameter short evacuated sleeve. The capacity of the whole insulated system is about 12 e.s.u. The Hoffman vacuum electrometer affords high charge sensitivity and great stability. In order to reduce the fluctuations in ionization readings caused by radioactivity of the ion-chamber walls the chamber was built 25 cm in diameter. The chamber walls were grounded and the saturation potential was applied to an internal grid of fine nickel wires just large enough to enclose the x-ray beam and the collector. Particles emitted from the walls thus fall short of the grid and the ions they produce do not reach the collector.[2] The insulator used was quartz. The chamber was filled with about an atmosphere of methyl bromide and the internal grid was long enough so that 98% of the energy of the x-radiation as hard as molybdenum K series could be absorbed. The window consisted of a thin slab of very light wood known as Balsa wood treated with paraffin to make it gas tight. It had quite a large opening about 2.5×8 cm. The inner surface of the window was coated with an extremely thin sheet of aluminum beryllium alloy 0.0002″ thick at the same potential as the grid. In this way, surface charges could not collect on the window and influence the collector. The entire system of ionization chamber and electrometer stood solidly fixed on a concrete slab. The ionization chamber and electrometer were standardized at frequent intervals with a radioactive source in a heavy lead case with a removable lead cover at a fixed distance of about a meter from the chamber. The radioactive source

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[2] We have since learned from A. H. Compton that he has previously built ionization chambers of this type.

was equivalent to 0.0547 mg of radium. It was necessary to use such a weak source so as not to disturb other workers in the laboratory. From time to time, the source was checked against a radium standard. By these precautions, it was possible to insure that intensity readings taken with the ionization chamber on different days might be strictly comparable. With this apparatus it has been found possible to reproduce single readings to within about 3600 ions per second. Probably not all of this uncertainty is ascribable to the ion chamber and electrometer. This is equivalent to an uncertainty of 5.6×10^{-16} amperes. Currents of 6×10^{-14} amperes can therefore be measured with 1 percent accuracy in a single observation. The authors hope to improve these figures by certain projected refinements.

The calcite crystals are mounted on pivots 12.5 cm apart *and are rotated simultaneously* through equal and opposite angles with respect to the line of centers of the pivots by means of two lever arms 43.5 cm long actuated one by a right the other by a left hand screw thread on one and the same shaft. A second similar mechanism geared to the first through a 2:1 gear ratio serves to rotate the x-ray tube source around the pivot of the first crystal at twice the rate of angular speed of the crystal and also to rotate the entire system of x-ray tube and spectrometer around a fixed pivot under and in line with the pivot of the second crystal so that the outgoing beam from the second crystal always remains lined up with the window of the ionization chamber. The entire mechanism permits the exploration of a range of two degrees for the *grazing angles* of incidence with the crystals without resetting the crystals. The smallest divisions on the drum correspond to 10 seconds of arc for the grazing angle and tenths of these divisions are easily estimated. All backlash greater than 15 sec. of arc is eliminated with helical steel springs. The entire apparatus is surprisingly simple to construct involving scarcely any very precise machine work and is very convenient to operate. Great precautions were taken at the start to insure the proper orientation of the crystal faces. These faces were accurately set at a definite known angle with the line joining the centers of their pivots and accurately parallel to their axes of rotation and as nearly as possible coincident with their axes of rotation by means of a spectrometer fitted with a Gauss eyepiece. The above described design makes it possible to explore a large range of wavelengths by turning a single drum without having to re-set the crystals. The mutual rotations of the elements of the apparatus *insure that the same parts of the surfaces of the two crystals are reflecting the x-rays at all times.* This eliminates errors due to (1) Crystals having different reflecting power in different parts of their surface. (2) Reduction of intensity due to part of the beam not falling on the crystal. (3) Changes in intensity due to the beam passing through different parts of the x-ray tube window or the ionization chamber window.

The x-ray tube was an ordinary water-cooled Coolidge type fitted with a copper target to which we have added a side window of mica 12×3 mm and about 0.0008 inches thick. Charcoal and liquid air are used to maintain a very good vacuum. We are much indebted to Samuel K. Allison for details of technique in making this window and also for his ingenious method of accurately controlling the filament heating current on the tube with a mercury column resistance. (Originally due to Paul Kirkpatrick).

A very convenient shielding box for enclosing the x-ray tube is made out of micarta tubing large enough in diameter to slip over the bulb and long enough completely to cover the tube. The micarta tube is wrapped outside with several layers of "leaded rubber" such as is sold by medical supply houses for the protective aprons of röntgenologists. A shielding box so constructed of insulating materials can be made much smaller and lighter than the large metallic lead boxes ordinarily employed. This is essential for the spectrograph here described since the tube in its shielding box must rotate around the pivot of the first crystal when the wave-length settings are being made.

The high tension generator was manufactured by the Wappler Co. and consists of a high tension transformer, two valve tube rectifiers and a mica condenser in oil. It has proved very stable in operation. A rough estimate of the ripple has been made for the voltage of excitation of the K lines of copper by the use of a Western Electric Braun tube oscillograph. The ripple is not more than 10%. The authors do not believe that the conclusions drawn from the results here described would be greatly affected by the complete elimination of this ripple. The voltage output to the tube has been carefully calibrated at 5.6 m.a. load from 6 to 14 kilovolts by measuring the current through about 2.5 meg ohms consisting of a number of large standard resistances in series. This curve was extended upward by means of an Abraham & Villard electrostatic voltmeter and a point at about 50 k.v. was checked with a measurement of the short wave-length limit with a Seemann Spectrograph. The short wave-length limit was also observed for the crystal setting at which $K\alpha_3$ was found and the voltage calibration could thus be checked accurately at this important point.

The radiation passed through two openings—one near the tube and one near the ion chamber—whose height limited the angle of "cross fire" through the spectrometer to 2.4° or 1.2° deviation from a plane normal to the edge of the dihedral angle formed by the two crystal faces.

The two crystals used were cleaved from the same block the faces previously in contact being used. The crystal was of unknown origin. It was obtained from Wm. Gaertner. The dimensions of the faces are 4.6×7 cm. These crystals are not of as high precision as those reported by some investigators; however, they give a line breadth at half maximum for the molybdenum lines of 0.4 X.U. The copper lines have a breadth at half maximum of 0.60 X.U. with these crystals.

METHOD

A spectral curve was first run for the $K\alpha_1$ and α_2 of copper in order accurately to establish fixed points on the scale of angle settings. This curve is shown in Fig. 1. It will be noted that the background appears absolutely flat at the point marked $K\alpha_3$ where the satellite should occur. The intensity of

$K\alpha_3$ in the case of copper is so low, that it is quite imperceptible plotted to this scale. In order to bring out the line, it was found necessary to take every precaution to reduce the background of the ionization curves. This background is doubtless due to four causes:

1. The continuous or "white" x-radiation. This is probably very small with the double crystal spectrometer since this instrument selects an extremely narrow band of wave-lengths. It was reduced by diminishing the vertical divergence of the beams in the spectrometer.

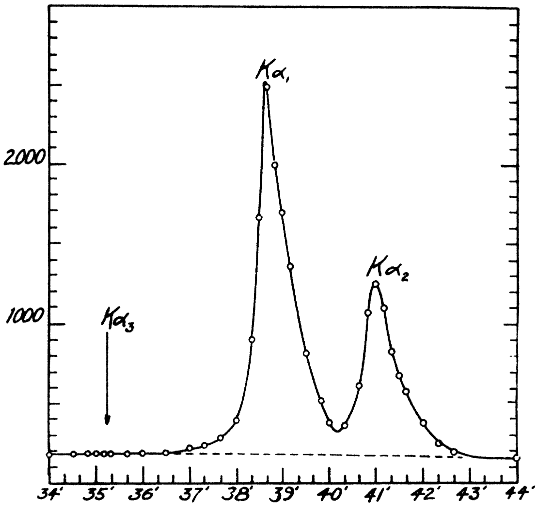

Fig. 1. $K\alpha_1$, α_2 of copper before taking precautions to reduce background in the neighborhood of α_3. Note that α_3 is quite imperceptible. The abscissae represent the grazing angles between x-ray beam and calcite crystals.

2. The natural leak of the electrometer and ion chamber. This has been reduced to an extremely low value by the precautions mentioned in describing the apparatus.

3. Nonselective scattering (Compton modified) from the crystal faces and fluorescent radiation from the crystals.

4. Direct leakage of x-rays from the tube to the chamber without selective reflection from the crystals.

It was found that the last two causes of background could be greatly reduced in the case of copper radiation by using a thin sheet of nickel as a filter to cut down the bulk of hard white radiation from the tube without greatly reducing the line intensity. The filter was a sheet of nickel rolled down to about 0.0005″ thick. Further reduction in the background was accomplished by great care in the arrangement of lead shields diaphragms etc. to preclude x-ray leakage and scattering to the highest possible degree and by reducing the vertical divergence of the x-ray beam through the spectrometer. We have succeeded in obtaining background ordinates of less than 1% of the maximum ordinate of $K\alpha_1$. See Fig. 2. This corresponds to only about double the spontaneous ionization observed by Millikan for this vicinity.

The procedure was then to run a spectral curve of $K\alpha_3$ holding the primary voltage and the tube current very accurately constant. It was found necessary to run all such curves after midnight to have the necessary constancy of the power supply. The attention of one observer was almost completely occupied holding the milliamperes constant as observed with a telescope and cross hairs focussed on the needle and keeping the primary voltage constant throughout the run. Time of drift of the electrometer was observed between two fixed points on the scale with a stopwatch. In general, only one or two curves could be taken in a night.

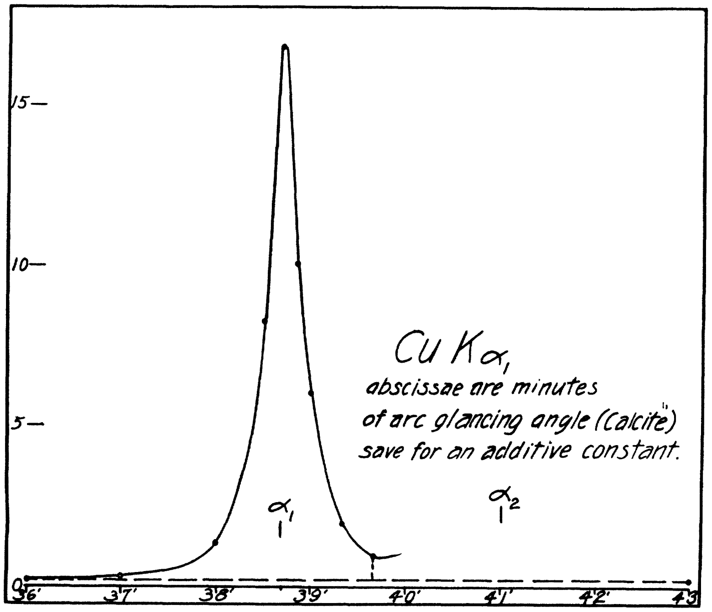

Fig. 2. Typical spectral curve of $K\alpha_1$ after taking full precautions to reduce background. From such curves as these the areas were taken for the purpose of determining α_1 intensities.

Although the background looks perfectly flat in the vicinity of $K\alpha_3$ in Fig. 1 when the sensitivity becomes sufficient to give good observations on $K\alpha_3$, the background is found to be increasing quite rapidly in this region. The "foot" of $K\alpha_1$ which is imperceptible relative to that line appears as a steep slope relative to the low intensity of $K\alpha_3$. Figs. 3 and 4 show the spectral curves taken on $K\alpha_3$ at various voltages all for the same current of 5.6 m.a. The enormous difference in intensity of the satellite and its parent will be appreciated when it is realized that the "saddle point" between α_1 and α_2 is about four times as high as the largest ordinates plotted in the curves of Fig. 3. The large background ordinates at 37′ in the curves of Fig. 3 are only about 1/50th(!) of the maximum ordinate of α_1.

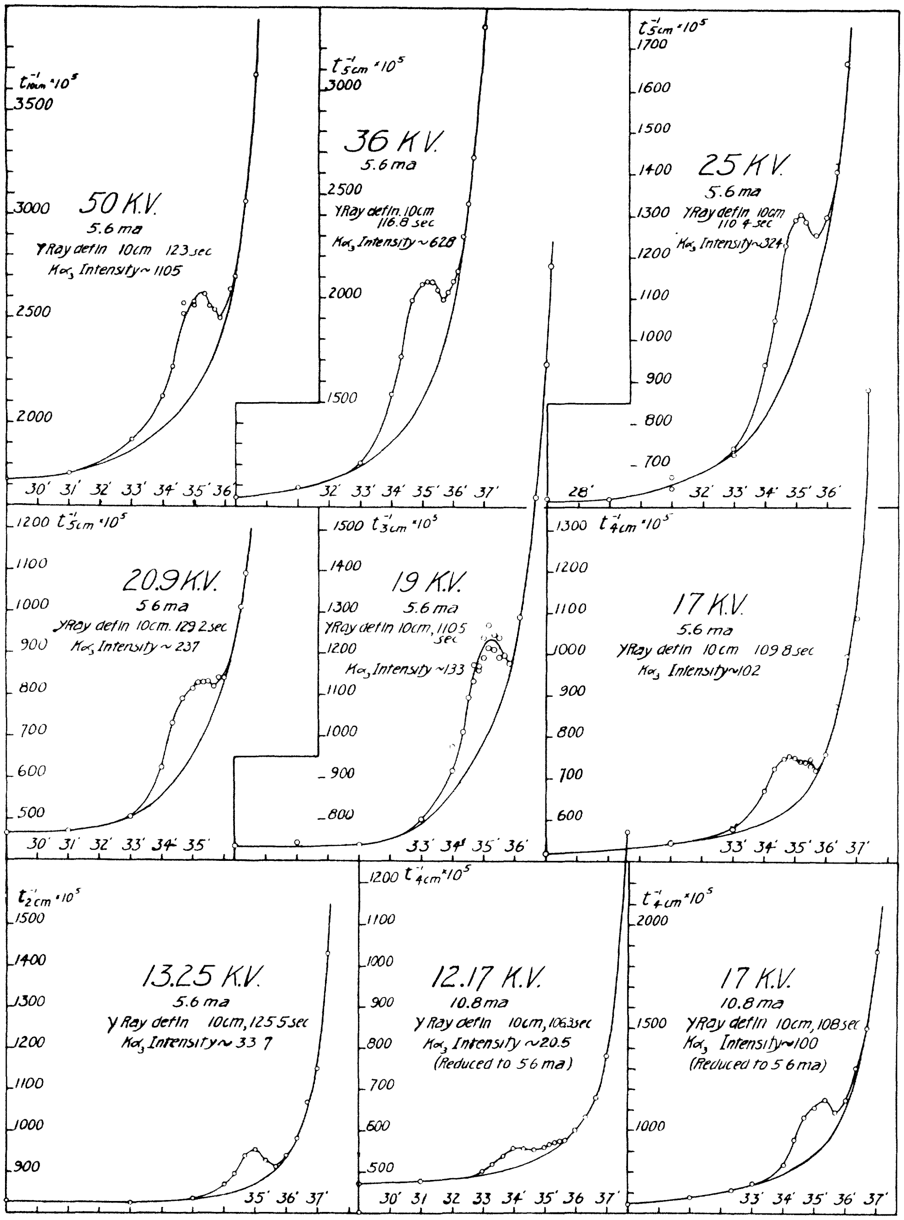

Fig. 3. Spectral curves of $K\alpha_3$. The area between the $K\alpha_3$ line and the interpolated background is taken as a measure of the intensity of the line. The sensitivity of the ion chamber and electrometer were calibrated in each case by taking the time for 10 cm deflection caused by γ rays from a radioactive source kept in a fixed position with respect to the apparatus. The calibrations are recorded on each curve shown. The ordinates of these curves are 10^5 times the reciprocals of the time in seconds required for a given number of cm deflection the number of cm being indicated as the subscript of the time. Note the partially resolved doublet structure which appears at the lower voltages.

The intensity of α_3 for the different voltages was obtained by interpolating the background curve under the region of α_3 as shown in the figure. This was done graphically with a spline. The areas of $K\alpha_3$ were then measured with a planimeter and checked by counting squares. These areas were then corrected according to the radioactive standardization of sensitivity for the time when the curve was taken. Not all the curves were observed for the same total deflection of the electrometer (in order to save time in the slow deflections) and this introduced another multiplying factor in the computation of the intensities from the measured areas. The different curves of α_3 are plotted to different ordinates scales introducing a third factor. The necessity of interpolating the background admittedly introduces an uncertainty in the measurements, but it is not possible to eliminate this.

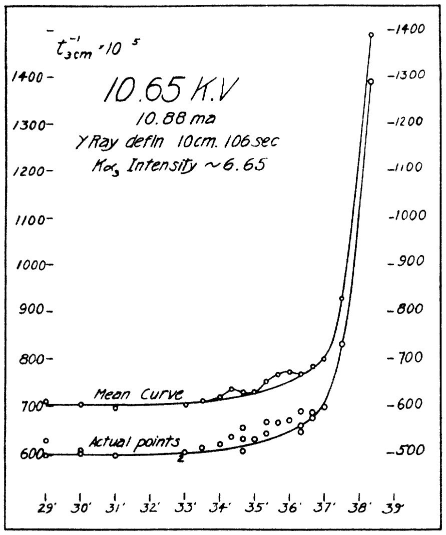

Fig. 4. Spectral curve of Cu $K\alpha_3$ at the lowest voltage where it was observed with certainty. Note the doublet structure. The actual observed points as well as the average curve are shown. The voltage here employed differs from the excitation voltage of the K series of copper by only 1.75 k.v.

From attempts to draw this interpolated background in different ways consistent with the background at considerable distance on either side of $K\alpha_3$ we have concluded that the uncertainty in area ascribable to this graphical interpolation is probably less than 5% of the total intensity in any case. Fig. 4 shows rather clearly a doublet structure in $K\alpha_3$ the separation of the components being about 2 X.U. Since it is the lowest intensity and voltage at which $K\alpha_3$ was observed with certainty, it is of especial interest. The voltage here differs from the excitation voltage of $K\alpha_1$ by only 1.75 k.v. Traces of the line were even thought to be detected at 9.75 k.v; but these results are rather uncertain.

A set of curves which it is not necessary to reproduce here was run for the line $K\alpha_1$ in the same way and for the same voltages and current as those shown in $K\alpha_3$.

RESULTS

Dependence of intensity on voltage. The intensities both of $K\alpha_3$ and $K\alpha_1$ are plotted (to different ordinate scales) as a function of the voltage squared in Fig. 5. It is seen that the intensities vary with voltage in almost identically the same manner. By projecting the nearly straight characteristic curves until they intersect the axis of abscissae the difference in excitation potential between $K\alpha_3$ and $K\alpha_1$ is estimated to be less than 200 volts. These projected intersections are not the excitation voltages themselves, but from the work of Wooten[3] it seems safe to assume that these intersections will serve to

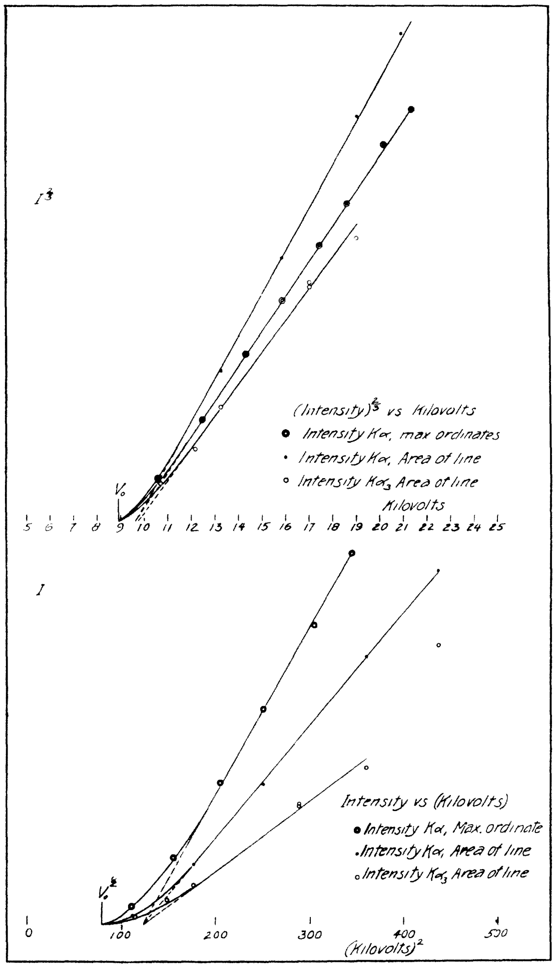

Fig. 5. Intensity of $K\alpha_1$ and $K\alpha_3$ as a function of voltage. These curves show how little difference (if any) exists between the excitation voltages of α_3 and α_1. The upper curve ($I^{2/3}$ vs k.v.) is plotted for Webster and Clark's relation $I=\kappa(V-V_0)^{3/2}$ while the lower curve (I vs. (k.v.)^2) is plotted for Wooten's relation $I=C(V^2-V^2_0)$.

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[3] Wooten, Phys. Rev. **13**, 71 (1919).

indicate any difference that might exist in the excitation voltages of $K\alpha_3$ and $K\alpha_1$.

The upper curve in Fig. 5 represents $I^{2/3}$vs. k.v. This method of plotting was adopted in order to rectify the curves in accordance with Webster and Clark's[4] observed intensity relation $I = K(V - V_0)^{3/2}$. It has the advantage of giving a smaller fillet in the region near the critical excitation voltage. This method of plotting also leads to the conclusion that $K\alpha_3$ differs in excitation voltage from $K\alpha_1$ if at all by less than 200 volts.

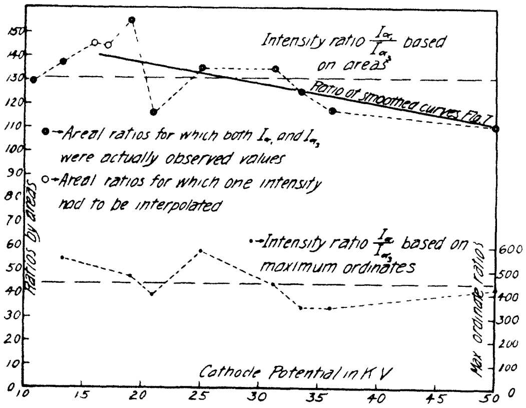

Fig. 6. Absolute ratio of $K\alpha_1$ to $K\alpha_3$ intensities from areas of spectral curves and also from maximum ordinates of each line at different voltages.

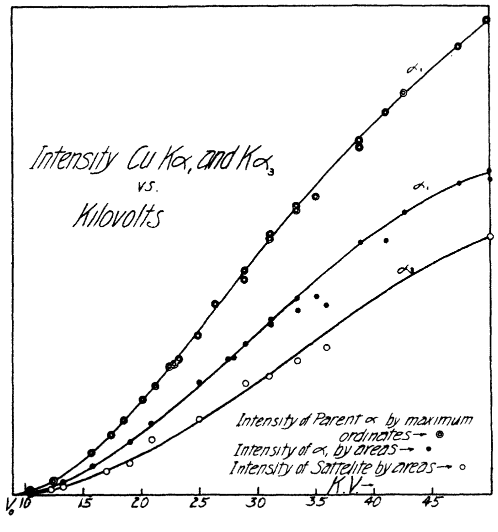

Fig. 7. Intensities of $K\alpha_3$ and $K\alpha_1$ for voltages up to five and one half times excitation voltage.

In Fig. 7 the intensities of $K\alpha_1$ by areas and ordinates and $K\alpha_3$ by areas

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[4] Webster and Clark. See Siegbahn's "Spectroscopy of X-rays," page 94.

are plotted up to 5.5 times excitation voltage. No very striking difference appears between α_1 and α_3. On the whole the ratio $I\alpha_1/I\alpha_3$ seems to increase slightly with decreasing voltage, see Fig. 6, down to about 15 k.v. This suggests a slightly higher excitation voltage for $K\alpha_3$ than for $K\alpha_1$.

Below 15 k.v. however the ratio appears to decrease again. The difficulties of measuring $K\alpha_3$ in this region make this decrease uncertain.

$K\alpha_3$ is 6X.U. harder than $K\alpha_1$. This corresponds to about 35 volts. If Richtmyer's "double jump" hypothesis is correct, this extra energy might be assumed to be contributed by a peripheral electron dropping to say an M level of about 35 volts. It should then require at least 35 volts or perhaps slightly more to ionize such an M level preparatory to such a transition. Since the target of the x-ray tube is a solid body it seems probable that the M electron would have to be ejected beyond the densely packed levels of the conduction electrons satisfying the Fermi statistics.[5,6] This would however require about 7 volts more. At the worst the Richtmyer "double jump" hypothesis could hardly require the $K\alpha_3$ line to be excited at voltages much more than 50 volts above the parent line, which is entirely consistent with the results reported in this paper.

The low intensity observed for $K\alpha_3$ of copper is also consistent with Richtmyer's hypothesis since the intensity of such a double transition depends on the "coupling" of the eigenfunctions for the four states involved which should be very much lower for copper than for the lighter elements in which $K\alpha_3$ appears more intensely.

2. Ratio of intensities. The ratio of intensities of $K\alpha_3$ to its parent $K\alpha_1$ based on areas is roughly constant for all voltages well above the excitation voltage and is found to be about 1:120. (See Fig. 6) The ratio of maximum ordinates of $K\alpha_1$ to $K\alpha_3$ is much larger than the ratio of areas because of the greater breadth of α_3 due to its doublet structure. This ratio is less accurately known but it is of the order of $I\alpha_1/I\alpha_3=440$.

3. Dependence of $K\alpha_3$ on current. This question was investigated by running two spectral curves of $K\alpha_3$ at the same voltage 17 k.v. one for 5.66 m.a. and one for 10.88 m.a. tube current. The conclusion from the measured areas is that the intensity of $K\alpha_3$ is accurately proportional to the first power of the current not to its square.

CONCLUSIONS

Although the difficulties and uncertainties of measurement of $K\alpha_3$ as a function of voltage caused by its extremely low intensity as compared to its parent line are admittedly considerable, it seems safe to conclude from this work that $K\alpha_3$ is excited at only very slightly higher voltages than $K\alpha_1$, the difference in excitation voltage being of the order of a few percent. It does not appear to us possible to explain the origin of $K\alpha_3$ of copper on the Wentzel-Druyvesteyn "spark line" theory. On the other hand, the results seem to be in accord with Richtmyer's "double jump" hypothesis.

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[5] A. Sommerfeld, Zeits. f. Physik **47,** 1–60 (1928).

[6] Jesse W. M. DuMond, Phys. Rev. **33,** 643 (1929).