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A NEW PRECISION X-RAY SPECTROMETER

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WALTER SOLLER

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Abstract

Multiple-deep-slit x-ray spectrometer.—A Bragg spectrometer was provided with two collimators, each consisting of a number of thin parallel strips of lead, separated a distance 1/166 times their length so as to produce many deep parallel slits. By this construction, radiation from a considerable area of the target passes through one or other of the slits of the first collimator, a large area of the specimen is thus irradiated, and also reflected radiation passing through one or other of the deep slits of the collimator fastened to the ionization chamber reaches a large area of cross section of that chamber; therefore the intensity of the ionization is relatively much greater than for the ordinary spectrometer. Any type of flat specimen can be studied, since in the large area irradiated properly oriented crystals are sure to be present, for some angles at least. The intensity curves obtained show sharp peaks, therefore lines can be accurately measured even when only 2′ of arc apart. Moreover, the large intensity available enables such peaks to be located for large angles between the axis of the collimators, increasing the accuracy. A test with a small sheet of steel gave accurate measurements of the MoKα_1 and Kα_2 peaks, the wave-length ratio coming out 1.00604 in agreement with Duane's value 1.00605. Also the expansion of the steel to 475°C was readily observed, the shift of 9′ giving a coefficient of 12×10^{-6}. The change in atomic distance produced by stretching a steel strip beyond its elastic limit was also measured, the contraction at an angle of 82° to the stress being 0.265 per cent. This instrument should prove useful in studying the effect of any agency tending to change the interatomic distances.

Introduction

TWO methods, the Bragg x-ray spectrometer and the Hull-Debye powder method, have been used for determining the atomic lattice patterns for a large number of crystalline substances. While these methods have given us very valuable information in regard to atomic structure, neither of them is adapted to determining quantitatively the interatomic changes due to thermal, mechanical and other physical and chemical influences on ordinary materials. For such work the apparatus described in this paper will be found to be superior to the former methods.

Apparatus

The electrical power was provided by two sixty cycle transformers connected to a mechanical rectifier and a filter circuit consisting of the secondaries of high tension transformers. A water cooled molybdenum

Coolidge tube was operated at a constant potential of 40 kv and a current of 30 m-amp. The rays reflected from the specimens were analysed with a Bragg x-ray spectrometer fitted with a Bumstead double tilted gold leaf electroscope.

The important modification of this spectrometer lay in the substitution of two specially designed collimators for the usual slits. The disposition of the apparatus and the construction of the collimators are shown in Fig. 1.

Each collimator consists of a large number of thin parallel strips of lead bound together as a unit in a tube to form a series of narrow rectangular spaces which are separated by very thin lead walls.

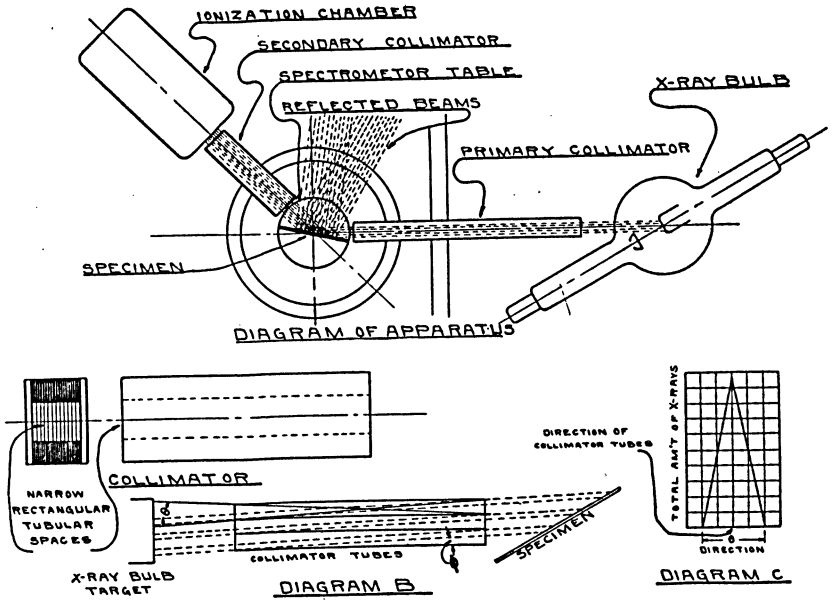


Fig. 1. Diagrams showing details of the spectrometer.

The first or primary collimator is placed between the specimen table of the spectrometer and the x-ray bulb so that the axis of the collimator is perpendicular to the face of the molybdenum target. The other or secondary collimator is fastened to the front of the ionization chamber and in line with it. The function of the primary collimator is to produce a wide but directionally restricted incident beam of x-rays; that of the secondary collimator is directionally to restrict the reflected rays which enter the ionization chamber.

The rectangular tubes of the primary collimator had a clear width of 1.24 mm and a length of 206 mm; those of the secondary collimator, a

width of 0.62 mm and a length of 103 mm. The wall thickness in each case was 0.08 mm. Because of the size of the ionization chamber, the cross-section of the secondary collimator was reduced to a 14 mm square.

The specimen was mounted on the spectrometer table with its plane face vertical and approximately in the line of the axis of rotation of the spectrometer. The angle between the incident beam of rays and the face of the specimen was established so that the reflected beam of rays was somewhat wider than the secondary collimator. The method of observing was the same as with other ionization x-ray spectrometers.

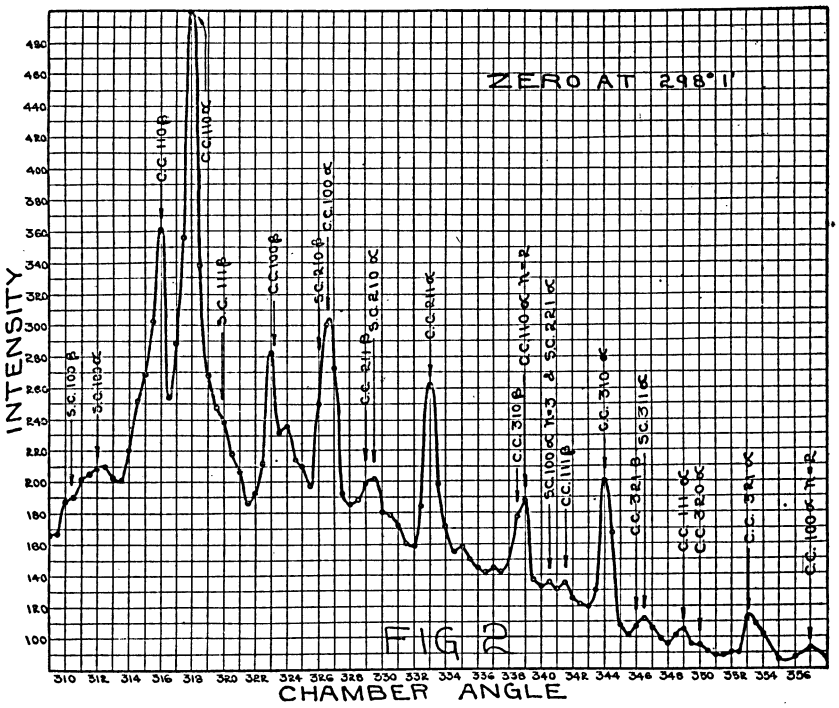
The primary collimator limits the direction of the rays in the incident beam to that included in the angle θ of diagram B, Fig. 1. All of the rays in the direction parallel to the axis of the collimator, are allowed to pass through the collimator. In the case of rays making an angle ϕ with the axis of the collimator, those that can pass through the collimator must be included in the series of small beams indicated by dotted lines in diagram B, Fig. 1. By plotting the cross-section of these beams with respect to the angle ϕ, through the range θ, a triangular plot of the amount of x-rays in the incident beam with respect to the direction of the rays (diagram C, Fig. 1) is obtained. This plot represents the characteristics of the collimator in selecting the rays according to direction. These characteristics of the collimator, as seen from the plot, give a pointed maximum of x-rays in the incident beam in the direction of the axis of the collimator. The sharpness of this maximum, and the limiting angle of divergence of the rays, is fixed by the ratio of the width to the length of the tubes of the collimator. As there are a large number of parallel tubes in the collimator, the incident beam will irradiate a large surface of the specimen, providing the area of the target emitting the x-rays is sufficiently large.

If the specimen is composed of a large number of crystals, variously orientated, each incident beam of homogeneous radiation will be reflected in a wide beam from every possible crystal plane of the specimen and there will be a pointed maximum for each of these beams. These sharp pointed maxima in each of the reflected beams will make an angle with the axis of the primary collimator in agreement with Bragg's equation.

The secondary collimator directionally analyzes the rays throughout these reflected beams. The characteristic curve of the collimator as an analyzer also has a pointed maximum. The plots of the ionization intensity readings with respect to angular setting of the ionization chamber, should therefore indicate the position of the reflected beams by peaks in the intensity curve and these peaks should have pointed maxima or crests. The position of the points of the crests should be a precise indication of the distance between the particular atomic planes giving rise to the beams.

Results

The first experiment was made with a plane strip of commercial steel subjected to the unscreened radiation of the molybdenum x-ray bulb. Readings of the ionization were taken at intervals of a half degree in the setting of the ionization chamber. The spectrum readings on the left side are shown in Fig. 2. The peaks formed chiefly by the reflection of the Kα doublet characteristic rays of molybdenum are marked α, and those for the reflection of the combined Kβ and Kγ rays are correspondingly



marked β. The designation *c.c.* 110α indicates the peak produced chiefly by the reflected α doublet characteristic x-rays from the crystal planes 110 (Miller indices), in a body-centered cubic atomic arrangement. The other combined peaks are marked according to the same scheme, where *s.c.* indicates a simple cubic arrangement.

Within the range of sufficient intensity, all the α and β peaks of a body-centered cubic arrangement are present and are prominent. Mingled with them are much smaller peaks usually corresponding to a simple cubic arrangement of atoms of the same elementary cube. Since the peaks for a body-centered cubic arrangement also correspond with peaks of a simple

cubic arrangement, only the additional peaks given by the simple cubic arrangement are designated by the letters *s.c.* Since all the peaks are present which should be expected from a body-centered cubic arrangement and are much the more prominent, we may consider this arrangement to be the typical one for the atoms of iron. This agrees with the opinion of Hull.

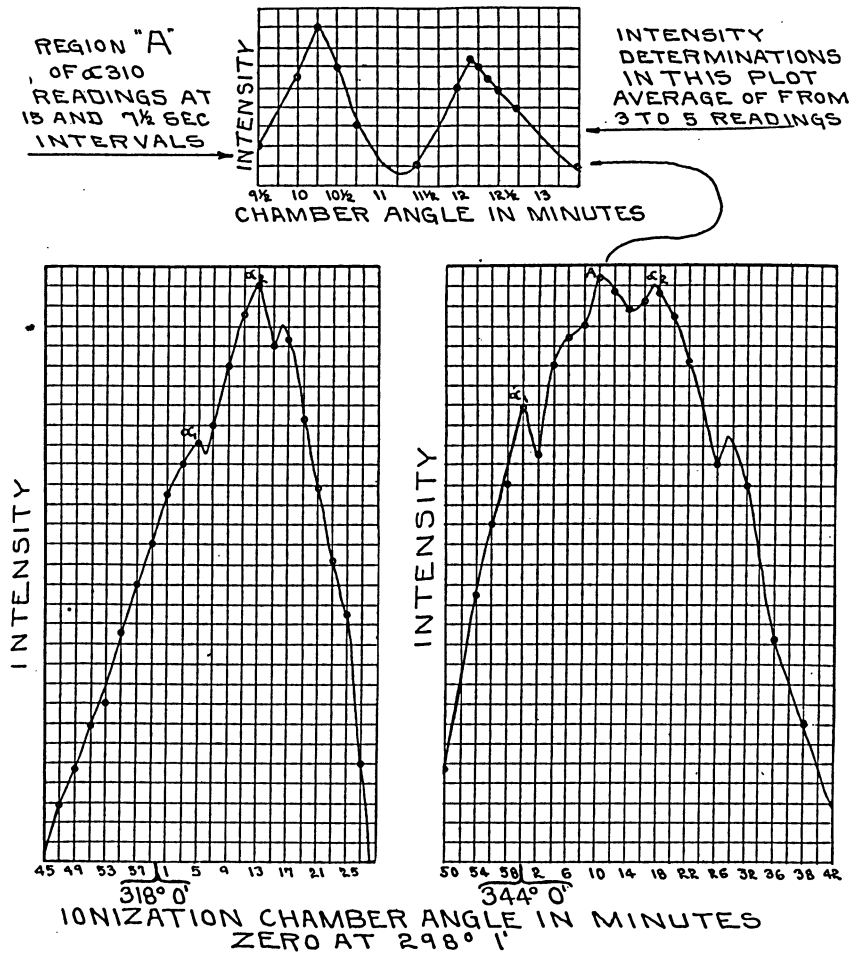


Fig. 3. Details of peaks *c.c.* 110*a* (left) and *c.c.* 310*a* (right).

To determine whether these peaks contained pointed crests, as the previous discussion indicated should exist, readings at two minute intervals of angle were taken in the *c.c.* 110*a* and *c.c.* 310*a* regions of Fig. 2. The plots of these small regions on a much magnified scale are shown in Fig. 3. The plot to the left is a magnified scale drawing of the portion from 317°45′ to 318°27′ (*c.c.* 110*a*) and the plot to the right is the analysis

of the region from 343°50′ to 344°42′ (*c.c.* 310α) of Fig. 2. The readings from 343°54′ to 344°22′ of the plot to the right in Fig. 3 were then checked and the existence of the component peaks in that part of the plot was confirmed.

It is evident that the apparently simple peaks are quite complex and in order to analyze still further their true contour, the region marked *A* Fig. 3 was examined more closely. Ionization readings were taken for each one quarter minute of arc and the single peak *A* broke up into the twin peaks shown in the small diagram, Fig. 3, separated by only 2′ of arc. It is safe to say that these peaks have been located to within $7\frac{1}{2}''$ of arc. The instrument evidently has a resolving power considerably greater than would be expected for triangular lines, which is $\frac{1}{2}\theta = 20'$.

The above results were confirmed by readings taken to the right of the incident beam. It was also shown that mechanical imperfections in the collimator were not producing extra peaks.

It is evident from these experiments that pointed crests are present in the plots of the ionization readings and that the position of the point of each of the crests can be located with great precision. It still remains to prove that the points of the crests, thus located, indicate precisely the angles which correspond to the correct values for a body centered cubic arrangement of the atoms and for the correct $K\alpha_1$ and $K\alpha_2$ characteristic wave-lengths of molybdenum.

According to Bragg's equation, the ratio of the distance between the 110 and the 310 planes is $\sin \theta_{310}/\sin \theta_{110}$. The present measurements give for the α_1 line, sin 23°8.2′/sin 10°6′ = 2.241, and for the α_2 line, sin 22°59.5′/sin 10°2′ = 2.242. The geometrical value of the ratio for a cubic crystal is 2.238. This agreement is much better than for measurements obtained by the powdered crystal method in which α_1 and α_2 peaks are not separated in the photographs.

Using the angles 22°59.5′ for α_1 (310) and 23°8.2′ for α_2 (310), the ratio of the corresponding wave-lengths is 1.00604, in accurate agreement with the ratio .71212/.70783 = 1.00605 as measured by Duane. This confirms our choice of the peaks which represent the α_1 and α_2 lines.

The peaks *A* and those at 318°16′ and at 344°28′ have not been definitely identified and as this has no bearing on the purpose of this investigation, no attempt was made to determine their significance.

As an example of the application of the method to the study of changes of structure due to physical changes, the effect of heating a small sheet of ordinary commercial steel was first investigated. This was used as the front wall of a small electric furnace, 7 cm long, 4 cm high, and 1 cm thick, heated by a flat coil of nichrome, embedded in fire-clay, and placed

so that there was only a very thin layer of clay between it and the steel specimen. A thermo-couple was soldered to the center of the face of the specimen where it would be irradiated by the x-rays. While the whole specimen was not heated uniformly by this method, the hottest part was located at the portion affected by the x-rays, and when it was heated to redness, an approximately uniform temperature was indicated by the uniformity of the color in this active region.

At the higher temperature the peaks were shifted to the left and decreased in intensity (see Fig. 4). By heating from 25° to 475°C, θ for the reflection from the *c.c.* 310 plane was 22°53′. This shifted from 23°2′ to

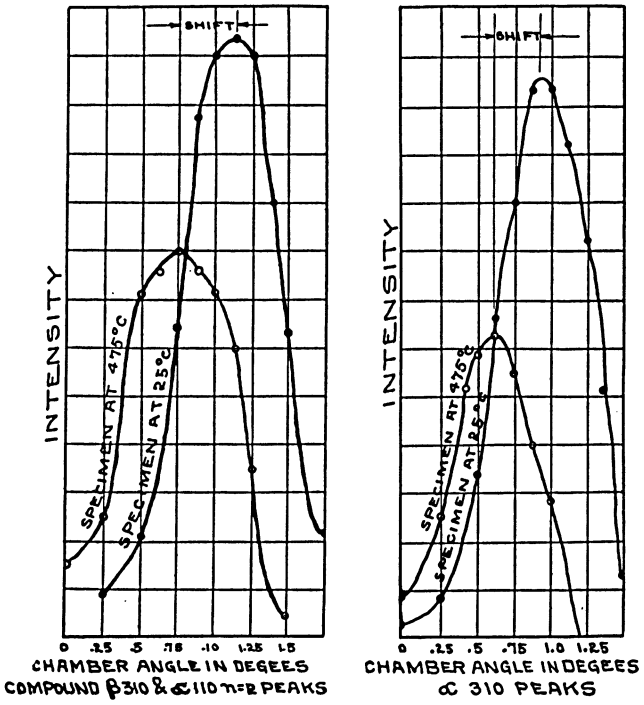


Fig. 4. Shift of peaks, due to expansion.

22°53′. This corresponds to an increase in grating space in the ratio 1.0055, giving a mean coefficient for the thermal expansion for steel of 0.122×10^{-4} in good agreement with other determinations. The agreement indicates that the method is reliable for the determination of interatomic thermal changes.

The decrease in the height of the peaks by increased temperature is in general agreement with the theoretical determination of Debye and the experiments of Bragg on crystals of rock salt. The flattening of the peaks

is very likely the result of the non-uniformity of the temperature of the specimen.

It is well known that metals strained mechanically beyond their elastic limit show variation in their granular structure as well as in their atomic distances. Since the former will not affect the x-ray spectrum, this method of analysis should be peculiarly valuable in distinguishing between these two effects. Since the angular range of the apparatus was limited by the particular collimators used, it was possible to determine only the decrease in atomic distances perpendicular to the direction of the force applied to the specimen, which is, by Poisson's ratio, approximately one-fourth of the corresponding increase in the atomic distances in the direction of the applied force.

A small sheet of steel, 16 mm wide and 0.18 mm thick from the same sheet as used in the previous experiments, was mounted in the tension apparatus, Fig. 5. By tightening the screws of this apparatus the specimen could be placed under a stress of 48×10^8 dynes per cm^2, which was just within the breaking load. The calibration of the instrument for this stress was correct within $\pm1\times10^8$ dynes. When the specimen was thus strained it was maintained for a day before readings were taken, to allow it to reach a steady state. The tension apparatus was placed on the spectrometer table at an angle of 15° with the incident beam and readings were taken at intervals of 2′ and 1′ of arc in the region of the *c.c.* 310α peak.

The shift of this *c.c.* 310α peak produced by this mechanical strain is shown graphically in Fig. 5. The contraction of the distance between the *c.c.* 310 planes under consideration is 0.0024×10^{-8} as calculated from the half shift 4′ and from θ for the unstrained specimen, $23°8\frac{1}{4}'$. The distance between these particular *c.c.* 310 planes of this strained specimen is thus $\lambda/2 \sin 23°12\frac{1}{4}' = (0.71212/.7880)\times10^{-8} = 0.9037\times10^{-8}$. The distance between the *c.c.* 310 planes of the unstrained specimen (previously calculated) is 0.9061×10^{-8}. Hence the contraction is 0.0024×10^{-8} or 0.265 percent. The angle between the normal to the *c.c.* 310 planes under consideration and the direction of the load on the specimen was 82°. The directly measured contraction of the specimen at 82° with the line of the load was approximately 4.5 percent.

Steel stressed beyond its elastic limit is in a plastic condition. In this condition it does not behave elastically as an isotropic body, as the crystalline nature of the grains then influences its elastic properties. The modulus of elasticity is less for stresses above than for those below the elastic limit. An indication of the decrease in the modulus is obtained by comparing the elastic strain obtained by this experiment, 0.265 percent,

with that value 0.07 percent that would have occurred had the stress of 48×10^8 dynes per sq. cm been below the elastic limit.

The significant information obtained from this experiment is that the shift of the 310α peak produced by a strain exceeding the elastic limit

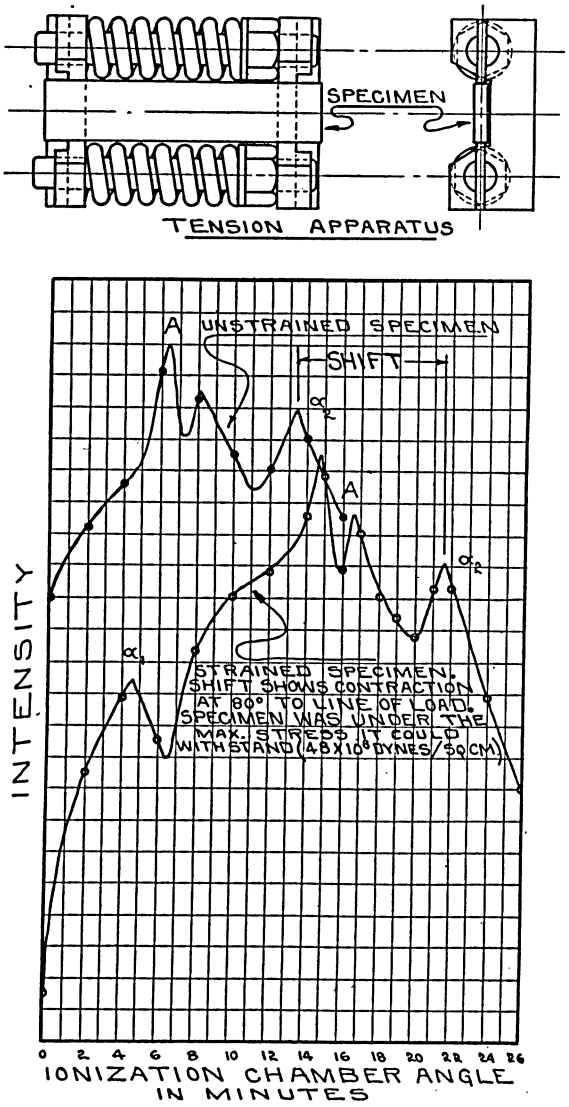


Fig. 5. Tension apparatus, and shift of 310α peak due to contraction produced by the stress.

of the specimen does not alter the shape of the curve or the relative positions of the secondary crests. Also, since the crests of the curve

remain sharply pointed, very accurate measurements of the shift can be obtained.

Besides the above experiments, the following facts were experimentally verified. First, the position of the pointed crests and therefore the accuracy of the atomic distance determinations are independent of the shape and condition of the specimen or the accuracy with which it is mounted on the spectrometer table. Second, the intensity of the reflected peaks increases approximately directly with the area of the specimen irradiated with x-rays, while the accuracy for any given intensity does not decrease with increase of the irradiated area of the specimen.

It should be noted that although the atomic distance determinations of these experiments are of higher accuracy than those obtained in previous work of this kind, the experiments are only preliminary. Even higher accuracy can be readily attained by improving the apparatus. A very considerable increase in the intensity of the peaks of the rays reflected from the specimen can be obtained by increasing the irradiated area of the specimen, the area of the face of the x-ray bulb target, and the energy output of the bulb. This increased intensity would permit the use of a smaller ratio of width to length of the collimator tubes, thus enabling more precise location of the point of a crest. This increase in intensity would also extend the angular range at which the position of the pointed peaks can be located and therefore the accuracy of the atomic distance determinations since the crests are better separated at the larger angles.

The chief advantage of this apparatus is that it makes it possible to analyze any kind of substance in the form in which it is produced, with high accuracy, and to determine the effect of any influence tending to change interatomic distances.

In conclusion I desire to express my thanks to the staff of the Physics Department, University of Cincinnati, and especially to Professor S. J. M. Allen, for assistance. I am also indebted to the Liebel Florsheim Co. for the gift of the x-ray equipment and to Mr. P. B. Evens for his very skillful construction of the special apparatus.

UNIVERSITY OF CINCINNATI,
March 21, 1924.