Second Series *January 1, 1932* *Vol. 39, No. 1*

THE

PHYSICAL REVIEW

X-RAY WAVE-LENGTHS BY THE DISPERSION IN QUARTZ

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(Received November 24, 1931)

Abstract

It has been shown that the quantum theory of dispersion accounts satisfactorily for the measured refraction of x-rays. Since the x-ray wave-lengths as determined by crystals and by ruled gratings differ by 0.25 percent, the dispersion may be used to indicate which of these values is correct. Thus the quantum theory of dispersion may be written

$$\lambda=\delta^{1/2}\left[\frac{\rho}{W}\cdot\frac{e}{m}\cdot\frac{F}{2\pi}\sum_{1}^{s}N_s\left\{1+\frac{\log(x^2-1)}{x^2}-\frac{2\pi q}{x^3}\right\}\right]^{-1/2}$$

where $\delta=1-\mu$, ρ the density, W the molecular weight, e/m the ratio of the electronic charge to mass, F the Faraday constant, N_s the number of electrons per molecule of frequency ν_s, $x=\nu/\nu_s$, ν the frequency of the incident radiation, $q=h/\nu_s$, k the damping factor which can be obtained from the atomic absorption coefficient. Precise measurements of the refraction of the copper and molybdenum K series in crystal quartz have been made. Both possible methods of using a prism were used. The 90° edge of the prism was very carefully prepared. The results obtained are given in the following table.

Method	$\delta\times10^6$	λ (Dispersion)	λ (Crystal)	λ(Grating)
1	8.553	1.536A	1.538A	1.542A
1	6.971	1.388	1.389	1.392
2	8.560	1.537	1.538	1.542
2	6.976	1.388	1.389	1.392
1 and 2	1.805	0.7089	0.7093	0.7109
1 and 2	1.432	0.6315	0.6314	0.6328

It is difficult to believe that such an agreement between the wave-lengths as determined by dispersion and by crystals is entirely fortuitous. The apparent precision obtained in the ruled grating measurements is so high that the above results must indicate a failure of the optical diffraction theory when applied to x-ray wave-lengths.

THE measured index of refraction of x-rays affords one of the most accurate tests of any theory of dispersion. Thus recent x-ray measurements[1] have shown that the quantum theory of dispersion as developed by Kronig,[2] Kramers, Kallman and Mark[3] more accurately accounts for the ob-

[1] A. Larsson, Inaugurald Dissertation, Uppsala (1929).
[2] R. de L. Kronig, Journal Opt. Soc. Am. **12**, 547 (1926).
[3] H. Kallman and H. Mark, Ann. d. Physik **82**, 585 (1927).

served values than does the classical theory. The well-known classical theory as developed by Drude and Lorentz may be written

$$\mu = 1 - \delta = \frac{e^2}{2\pi m} \sum_1^s \frac{n_s}{\nu^2{}_s - \nu^2} \tag{1}$$

or

$$\frac{\delta}{\lambda^2} = \frac{\rho}{W} \cdot \frac{e}{m} \cdot \frac{F}{2\pi} \sum_1^s \frac{N_s}{\lambda_s{}^2 - 1/\lambda^2} \tag{2}$$

where μ is the index of refraction, e the charge on the electron, m the mass, n_s the number of electrons per unit volume of natural frequency ν_s or wave-length λ_s, ν the frequency or λ the wave-length of the incident x-rays, ρ the density of the refracting medium, W the molecular weight, F the Faraday constant, and N_s the number of electrons per molecule of frequency ν_s. Eq. (2) has been used[4] to determine the value of e/m from the measured value of δ. Since there is now very little question about the real value of e/m we can get some information concerning the real wave-length of the x-rays λ. Thus Eq. (2) may be solved for λ

$$\lambda = \delta^{1/2} \left(\frac{\rho}{W} \cdot \frac{e}{m} \cdot \frac{E}{2\pi} \sum_1^s \frac{N_s}{\lambda_s{}^2 - 1/\lambda^2} \right)^{-1/2}. \tag{3}$$

The approximate values of λ and λ_s are known. It has been shown that the x-ray wave-lengths obtained by using crystal gratings differ by about 0.25 percent from those obtained by using ruled gratings. While Eq. (3) cannot be expected to give precise wave-lengths, it should indicate which of the two existing values is correct. It was pointed out above that Eq. (1) does not agree as well with the experimental facts as does the quantum theory, but it was thought best to indicate the method of determining λ in this simpler equation, although all later calculations have been made using the quantum theory.

The quantum theory has been put in a very convenient form by Prins.[5] The index of refraction may be written

$$\mu = 1 - \sum_1^s \alpha_s - \sum_1^s \beta_s i \tag{4}$$

where α and β are given by:

For the region $\nu_s > \nu$

$$\alpha_k = \frac{e^2}{2\pi m} \cdot \frac{n_k}{\nu^2} \left[1 + \frac{\log (1 - x^2)}{x^2} \right] \tag{5}$$

$$\beta_k = \frac{e^2}{2\pi m} \cdot \frac{n_k \nu_k}{\nu^3} \left[\frac{2 - x^2}{1 - x^2} + \frac{2 \log (1 - x^2)}{x^2} \right]. \tag{6}$$

[4] J. A. Bearden, Phys. Rev. **38,** 835 (1931).
[5] J. A. Prins, Zeits. f. Physik **47,** 479 (1928).

For the region $\nu > \nu_s$

$$\alpha_k = \frac{e^2}{2\pi m}\cdot\frac{n_k}{\nu^2}\left[1 + \frac{\log (x^2 - 1)}{x^2} - \frac{2\pi q}{x^2}\right] \tag{7}$$

$$\beta_k = \frac{e^2}{2\pi m}\cdot\frac{n_k}{\nu_k^2}\left[\left\{\frac{x^2 - 2}{x^2 - 1} + \frac{2 \log (x^2 - 1)}{x^2}\right\}\frac{q}{x^3} + \frac{\pi}{4}\right] \tag{8}$$

where $x = \nu/\nu_k$, q is the damping factor divided by ν_k and e and m are the same as in Eq. (1). The $\sum_1^s \alpha_s$ corresponds to the δ used in Eqs. (1), (2) and (3), and the imaginary term $\sum_1^s \beta_s i$ is proportional to the absorption of the x-rays. The value of q in Eq. (7) can be obtained from Eq. (8) by substituting for β its value as obtained from the measured atomic absorption coefficient. Since we are primarily interested in the region $\nu > \nu_s$ we can obtain from Eqs. (4) and (7) a value of λ similar in form to that given in Eq. (3) or

$$\lambda = \delta^{1/2}\left[\frac{\rho}{W}\cdot\frac{e}{m}\cdot\frac{F}{2\pi}\ \sum_1^s N_s\left\{1 + \frac{\log (x^2 - 1)}{x^2} - \frac{2\pi q}{x^3}\right\}\right]^{-1/2} \tag{9}$$

Experiment

There are three methods by which the index of refraction of x-rays may be measured. First, the deviation from the Bragg law, second the total re-

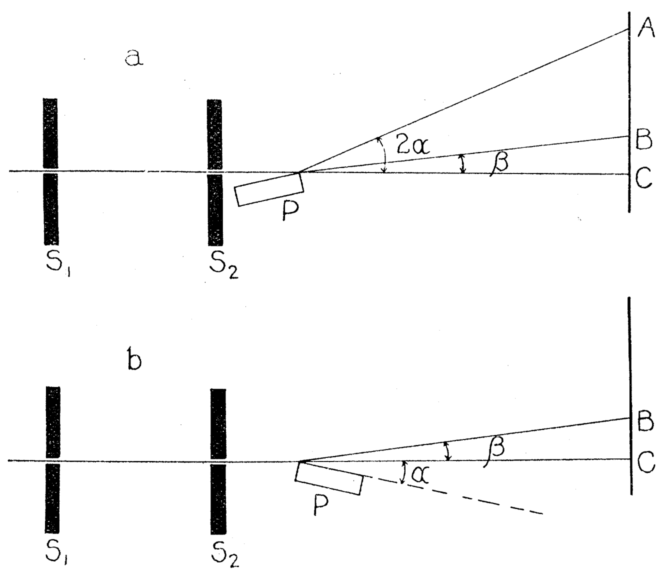

Fig. 1. Apparatus. Slits S_1 and S_2 were 0.015 mm wide and 40 cm apart. The distance from the prism P to the plate was about 200 cm.

flection of x-rays, and third the prism method. Since the prism method is the most direct and precise, it has been used in the present experiments.

The two methods of using a prism for determining the index of refraction can be easily explained by reference to Fig. 1a and b. In Fig. 1a the x-ray beam struck the first face of the 90° prism at a glancing angle slightly larger than the critical angle of total reflection for the copper $K\alpha$ line. The continuous spectrum of longer wave-length was reflected from this face of the prism and was used to determine the angle between the direction of the x-ray beam and the surface of the prism. The reflected beam was recorded on a photographic plate at A, at the same time the refracted beam was recorded at B. The direct beam was recorded at C. Thus the angles α and β needed for determining μ can be obtained from the separations of the lines on the photographic plate, and the distance R from the edge of the prism to the plate. The index of refraction μ can be calculated from the equation

$$\mu = 1 - \delta = \frac{\cos \alpha}{\cos (\alpha - \beta)} \tag{10}$$

and for small angles α and β this may be written (11)

$$\delta = \beta(\alpha - \beta/2).$$

In the second method (Fig. 1b) the x-ray beam struck the second face internally at almost zero glancing angle and left at nearly the critical angle of total reflection. Stauss[6] has shown that the refracted beam should be less divergent than the incident beam in this case, and all wave-lengths should suffer maximum deviation simultaneously. This method, however, necessitates the precise measurement of a small angular rotation of the prism in order to determine the angle α. It is obvious from Fig. 1b that δ can be obtained from Eqs. (10) and (11).

Since the absorption of the x-rays in the prism limits the useful portion to a region very close to the edge of the prism it is very necessary that the edge be carefully prepared. In order to obtain as sharp an edge as possible a crystal quartz plate 33×65 mm was polished optically flat and then cut into two pieces 33×32 mm. These pieces were cleaned and worked together until the central white light interference fringe showed that the two polished surfaces were in practically perfect contact. These pieces were then placed in a rigid clamp, and the edges which were produced when the large plate was cut into were ground off until the line of demarcation between the plates practically disappeared. This edge was then polished and when the plates were separated a microscopic examination of the 90° edge showed it to be very sharp.

The precise adjustments of the slits, prism and plate holder were made by the method described by the writer[7] in the measurements of x-ray wavelengths by ruled grating. The location of the edge of the prism on the axis of rotation of the prism table was made with the aid of a Michelson interferometer. The slits were about 0.015 mm wide and 40 cm apart. The distance of the plate holder from the edge of the prism was about 2 meters. The source of x-rays was a water-cooled copper or molybdenum target Coolidge tube

[6] H. E. Stauss, Phys. Rev. **36**, 1101 (1930).
[7] J. A. Bearden, Phys. Rev. **37**, 1217 (1931).

with a small focal spot. The tube was operated at about 10 m.a. and 30 kv peak. In order to reduce the exposure time to a minimum, a simple calculation shows what fraction of the primary beam should be absorbed. The adjustment of the prism to absorb this fraction was easily made with the aid of an ionization chamber.

The exposures varied from 30 min. to three hours, depending upon the angular deviation of the refracted beam. Eastman x-ray plates were used for recording the positions of the various x-ray lines. The separations of these lines were measured with a calibrated comparator. Measurements were made on both the unresolved $K\alpha$ doublets and the $K\beta$ lines. The average wave-length of the $K\alpha$ doublet was obtained from the equation

$$\lambda = \tfrac{1}{3}(2\lambda_1 + \lambda_2)$$

where λ_1 is the wave-length of the $K\alpha_1$ line and λ_2 the $K\alpha_2$ line. The wave-lengths as obtained with crystal gratings are

copper $K\alpha = 1.538\text{A}$	molybdenum $K\alpha = 0.7093\text{A}$
copper $K\beta = 1.389$	molybdenum $K\beta = 0.6314$

The corresponding wave-lengths as obtained from ruled gratings[7] are

copper $K\alpha = 1.542\text{A}$	molybdenum $K\alpha = 0.7109$
copper $K\beta = 1.392$	molybdenum $K\beta = 0.6328$

The density of the quartz prism was determined by the method described in a recent issue of this Journal.[8] The density was

$$\rho = 2.6485 \pm 0.0002 \text{ gr/cm}^3.$$

Results

The first measurements[4] were made with the prism as shown in Fig. 1a using the copper K radiation. The angle α was varied from 16′30″ to 27′57″ in the different exposures. Eighteen measurable exposures were obtained by this method. Fig. 2a shows a typical plate. The results were

$$\delta_\alpha = 8.553 \pm 0.006 \times 10^{-6}$$
$$\delta_\beta = 6.971 \pm 0.005 \times 10^{-6}.$$

The probable error was calculated from the variations in the results by the method of least squares. The alignment of the apparatus and the measurement of the distance from the edge of the prism to the plate holder have been so carefully made that the writer believes the maximum error in δ from these sources is less than $\pm 0.002 \times 10^{-6}$. It should be pointed out, however, that an indeterminate error may arise from the fact that it is necessary to measure to the edge of the reflected beam. The length of the exposures was varied over a wide range in order to reduce this possible error to a minimum. The agreement between the present results and those obtained by Larsson[1] indicate that this error must be negligible.

[8] J. A. Bearden, Phys. Rev. **38,** 2089 (1931).

The next measurements were made with the some wave-length as above but with the prism as shown in Fig. 1b. This method should have given a refracted line narrower than was obtained in the first method.[6] A comparison

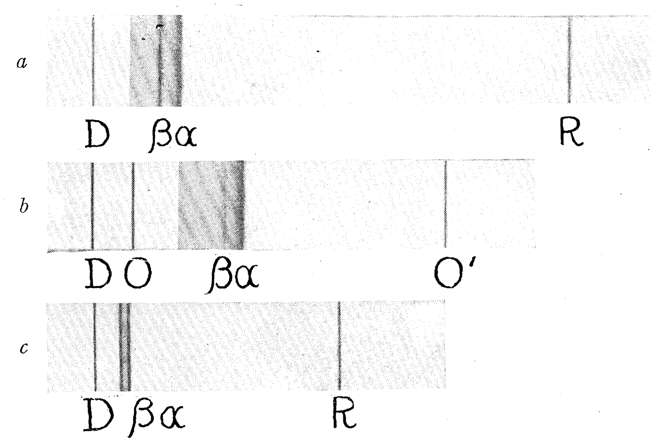

Fig. 2. Three-fold enlargements of typical plates. D is the direct beam, β and α are the refracted $K\beta$ and $K\alpha$ lines, R is the reflected beam, O and O' are reflected lines used for determining the angles with method 2.

of a typical plate shown in Fig. 2b with the plate in Fig. 2a shows that there is very little if any difference in the widths of the refracted lines. The results from 11 plates taken under various conditions gave

$$\delta_\alpha = 8.560 \pm 0.008 \times 10^{-6}$$
$$\delta_\beta = 6.976 \pm 0.007 \times 10^{-6}$$

where the probable error was calculated as above.

After the above results had been obtained, the copper target x-ray tube was replaced by a molybdenum tube and a series of measurements similar to those above were made. Fig. 2c shows a typical photograph. The deviation of the refracted ray from the direct beam was much less in this case than above, but appeared sharper. Thus the results are just about as precise as those for the copper radiation. The average of all the results obtained with the molybdenum K series gives

$$\delta_\alpha = 1.805 \pm 0.001 \times 10^{-6}$$
$$\delta_\beta = 1.432 \pm 0.001 \times 10^{-6}.$$

Calculations of Wave-Lengths and Discussion

As was pointed out above, the bracket term of Eq. (9) is a constant for slight variations of λ. Thus the wave-lengths used in this term may be changed by ± 0.25 percent and the value of the bracket will be changed by less than one part in 5000. The results given in Table I have been calculated assuming wave-lengths which were the average of the wave-lengths as given

TABLE I.

Method	$\delta \times 10^{-6}$	λ (Dispersion)	λ (Crystal)	λ (Grating)
1	8.553	1.536A	1.538A	1.542A
1	6.971	1.388	1.389	1.392
2	8.560	1.537	1.538	1.542
2	6.976	1.388	1.389	1.392
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1 and 2	1.432	0.6315	0.6314	0.6328

by the crystal and grating methods. The wave-lengths calculated from Eq. (9) with the measured δ are given in the column headed λ (dispersion). The agreements between the wave-lengths as obtained from dispersion and those with crystal gratings are not perfect, but it appears to the writer that the agreement is entirely too close to be fortuitous. Larsson[1] has also reported seven determinations of δ for the copper $K\alpha$ line refracted by a quartz prism using essentially method 1. His results when used as above give

$$\lambda = 1.536\text{A}$$

which is in exact agreement with the writer's result for method 1, Table I. Stauss[6] has measured δ for the molybdenum $K\alpha_1$ and $K\beta$ lines in quartz by using method 1. His results are slightly higher than those obtained by the writer and give

$$\lambda_\alpha = 0.7087\text{A}$$

$$\lambda_\beta = 0.6325$$

which are in some better agreement with the grating wave-lengths than the crystal values. Stauss[6] apparently did not take into account the natural width of the refracted beam and this may account for the difference of his results and those obtained by the writer.

These results thus indicate that the grating measurements of x-ray wave-lengths are definitely in error. Considering the precautions taken in the grating measurements[7] and the general agreement between various observers, the cause of the difference in the two methods must be due to a failure of the ruled grating diffraction theory when applied to x-ray wave-lengths.

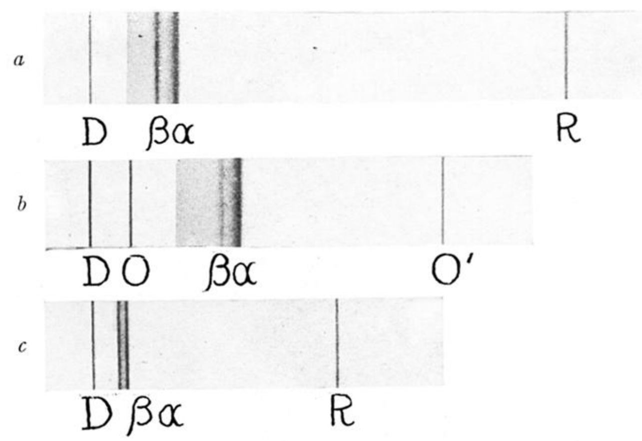

Fig. 2. Three-fold enlargements of typical plates. D is the direct beam, β and α are the refracted $K\beta$ and $K\alpha$ lines, R is the reflected beam, O and O' are reflected lines used for determining the angles with method 2.