Second Series *April, 1928* *Vol. 31, No. 4*

THE

PHYSICAL REVIEW

THE REFLECTION OF X-RAYS BY PLANE SURFACES

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Abstract

The reflection of the $K\alpha_1$ radiation of molybdenum by plane surfaces of glass and quartz and by sputtered films of nickel, platinum, and silver has been investigated. It was found that the critical angle of reflection of nickel was smaller for transparent films than for opaque ones. The critical angles for both nickel and platinum were lower than the values calculated from the electron theory. The experimental value for the critical angle of silver agreed with the theoretical within the limits of error of the experiment. Doan has found experimental values of the critical angle for films of a number of metals that agree with those predicted by the theory, and Edwards has obtained similar results using solid reflectors. If it is to be granted that the theory does hold true in the case of nickel and platinum, too, it is necessary to assume that the densities of sputtered films of nickel and platinum are less than those of the metals. Further it must be assumed that in the case of nickel the density varies with the thickness of the film, being smaller for the thinner films.

The observed critical angle for quartz was smaller than the theoretical value, as though the surface density was less than the average density. Both glass and quartz surfaces showed a weak residual reflection beyond the critical angles, of a type that was not present in the sputtered films. The phenomenon may be caused by irregular densities over the surface due possibly to the polishing or to the heterogeneous composition of the two substances.

The Experimental Method

THE experiments described in this paper were performed for the purpose of studying the reflection of x-rays by plane surfaces. The general principles underlying the investigation are well presented in other papers,[1,2,3] and need not be discussed here. The experimental method used by the author was, in brief, to split a beam of homogeneous x-rays with the mirror itself, causing part to be reflected and allowing part to pass by undeviated, and to determine the angle of reflection by measuring the separation of the two resulting beams on a photographic film. Fig. 1, which is not drawn to scale, shows the arrangement of the apparatus. The x-rays from the target A are reflected by the calcite crystal B, and are split by the mirror D. No slits were used except to prevent undesirable radiation from getting into the room. A narrow beam was secured by taking the radiation from the target of a water-cooled tube at a glancing angle of 14°; the crystal was relied upon to

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[1] R. L. Doan, Phil. Mag. **4**, 100 (1927).

[2] H. W. Edwards, Phys. Rev. **30**, 91 (1927).

[3] H. E. Stauss, Nature **114**, 88 (1924).

limit the beam of monochromatic radiation that was used to a small angular width. The edge of the mirror farther from the x-ray tube was over the center of rotation of the spectrometer and did not move appreciably during the rotation of the mirror. The edges of the beams used in the measurements were those determined by this farther edge of the mirror, and were as well defined as if they had been limited by a slit. The distance from crystal to mirror edge was 30 cm. The $K\alpha_1$ radiation of molybdenum, of 0.7076A wave-length, was used throughout the work.

The experimental procedure involved turning the mirror through successively increased angles and making ten minute exposures in each position. The pictures were taken at a distance of 98.1 cm from the mirror edge until the reflected beam was invisible; then the distance was shortened to about 11 cm. In the new position of the film the reflected line became visible again, and the procedure for the longer distance was continued. The angles of reflection could be determined directly from measurements of the films at the greater distance until the reflected line became too weak to be measured accurately.

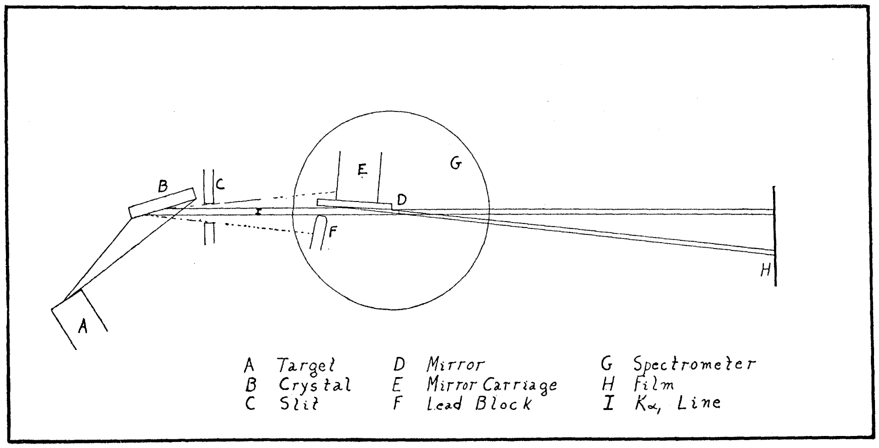

Fig. 1. Experimental arrangement.

For these larger angles and for those with the film at the shorter distance the angular displacements of the reflector from a standard position, as measured by a sensitive optical system, were used to determine the angles of reflection. The sensitiveness of the system depended upon the use of monochromatic light and of a low-power microscope to read the deflections of the image. The source was a filament of a tungsten red-lamp, such as is used in dark-rooms. The real image of the filament was observed directly by the microscope at a distance of 181.0 cms from the mirror. A collodion color filter was placed over the objective of the microscope. It, together with the red glass of the lamp, permitted only a very narrow band of the spectrum in the extreme red to reach the microscope. An extremely sharp image was formed, whose edge could be read absolutely to 0.02 mm. Small angular displacements of the mirror could be read with a probable error of less than 0.2″.

The relative intensity of the reflected to the primary beam was found by comparing the relative blackening of the two visually. As both lines always appeared on the same films, this method consisted merely of a simple comparison of the densities of two lines produced under the same conditions. The relative values of the densities at the two different distances were obtained by taking pictures for one angle in both positions in each series and the results were checked by calculations based on the geometry of the apparatus. More accurate values of the relative intensities were not necessary since the final angle of reflection is the important datum. The results were plotted and are shown in Fig. 2. It should be noted that all the angles in the graphs are measured relative to a standard position with the optical system—the standard position for each trial being one of the angles at which the reflected line was strong.

In this method the error made in measuring any angle of reflection is the error made in determining the standard position added to the error made in reading the displacements from it. The second error is negligible compared to the first and may be neglected. The error made in measuring the reference angle is half the angular width of the beam, if the effect of the penumbra of the lines on the measurements is neglected. The angular spread of the primary beam in this experiment, 43′′, was determined by measuring its width at two different positions. Actually, however, a penumbra occurs on each edge of the lines within which the intensity varies from zero on the outside to the full intensity of the beam. Fig. 3 illustrates the effect, where both a_1b_1 and a_2b_2 are penumbrae. It also illustrates the error in the determination of the angle of reflection, if the effect of the penumbra is neglected; a_1b_2 is the distance that would be measured, while b_1b_2 is the desired distance. As all readings must be made from somewhere within this penumbra, an error is introduced into the result which is indeterminable. If all readings are assumed to have been made in the region of zero to half intensity, as seems likely to be the case, the error made in determining an angle lies between

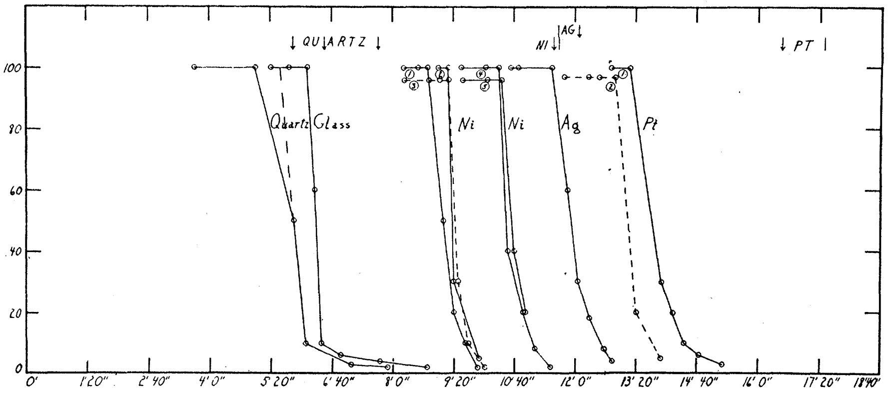

Fig. 2. The relative intensity of the reflected to the primary line plotted against angle of reflection.

one-quarter and three-quarters of the width of the beam, or between 11″ and 33″. The actual error is probably somewhat larger, however, because the effect of the penumbra upon the determination of the spread of the beam has been neglected. The measured values of the angles will always be greater than the true values.

The critical angle of reflection that is desired is the final angle of reflection, which was apparently increased in this method by the curvature of the mirror, but the error was only 4″ and could be corrected for. An examination of the curves shows that the drop in relative intensity of reflection between the value of 100 and 20 (the limit of visibility at the greater distance) does cover approximately 50″ as would be expected, although the total drop including that at the shorter distance, occurs through an appreciably larger angle. If this additional reflection is due to very weak radiation with more than 43″ spread, it may be neglected and the critical angle assumed to be the limit of visibility at 98.1 cm, or a relative intensity of 20 for the two beams. The true value of this angle will then be 15″ to 37″ less than the measured angle. In view of the uncertainties of the corrections to be applied, all of the angles used in the graph are uncorrected.

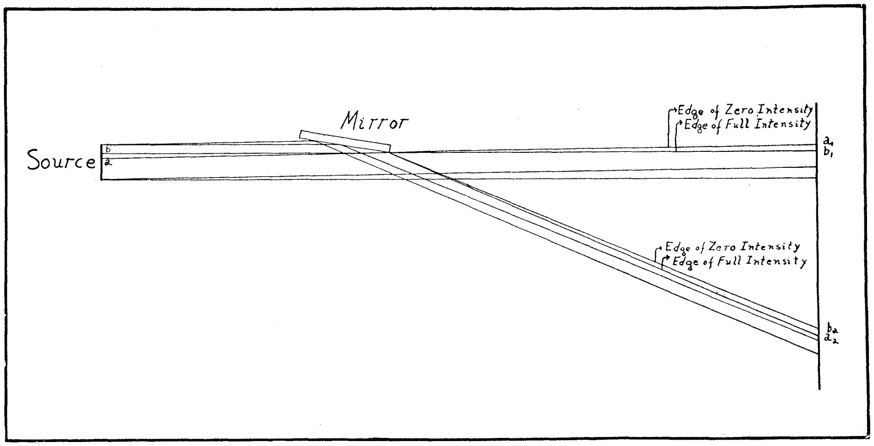

Fig. 3. A diagram illustrating the effect of the penumbra upon the measurements. For convenience in drawing, the crystal has been used as the "source" of the radiation (see Fig. 1). The beam that comes from it, if this is one of the characteristic radiations, has a definite angular width without the use of any slits. In the sketch this angular width is the angle subtended by ab, a_1b_1 or a_2b_2 at the mirror. The drawing differs from that in which the source emits radiation in all directions in that the limiting rays are defined by the "source" (or crystal) instead of by slits. For example, no rays outside the length of ab of the crystal can strike the mirror edge, because that would require a greater angular width of the beam than is found experimentally.

RESULTS

The substances investigated were glass, quartz, and films of silver, nickel, and platinum sputtered on the glass surface. In the case of nickel it was discovered that the critical angle depended upon the thickness of the film.

Curves 1 and 2, Fig. 3, represent the reflection from two transparent films sputtered in hydrogen, and curve 3 that from one sputtered in nitrogen Curves 4 and 5 represent the reflection from the same opaque film of nickel sputtered in hydrogen. The difference between the two sets of curves is about 80″, more than can be attributed to experimental error. The phenomenon can be explained easily if it is assumed that the thin films are less dense than the thick ones. This is in accord with the results of J. D. Hanawalt and L. R. Ingersoll[4] who found that nickel films sputtered in hydrogen are less dense than the metal itself. However, to explain the present case, it must be assumed that the low density is not due to the presence of hydrogen in the film, but to the thinness of the film or to the presence of any gas in it. If Lorentz's theoretical formula for the critical angle of reflection is true, the difference in density of the two types of films is about 23 percent of the more dense one.

If, further, the critical angles are compared with the theoretical values, it is found that they are much smaller than can be accounted for by experimental error in the case of both nickel and platinum. The theoretical critical values are shown in the upper part of the graph, the arrows indicating those obtained by neglecting the effect of the critical frequencies and the straight lines those found by assuming these to be the absorption frequencies and using Stoner's distribution of electrons. Curve 1 for platinum represents the reflection for an opaque film sputtered in hydrogen and curve 2 for one sputtered in nitrogen. If the discrepancy between experimental and theoretical values is to be explained as being due to density changes, both the nickel and platinum films, even those that are opaque, must have lower densities than the metals themselves. Considering Lorentz's formula as true again, the density of the platinum is only about 75 percent of that of the metal.

The following table shows the approximate densities of the films, if Lorentz's formula is assumed to hold.

Element	Nature of Film	Density of Element (gms/cc)	Density of Film (gms/cc)
Ni	Transparent	8.90	4.13
	Opaque	8.90	5.44
Pt	Opaque	21.37	14.1

The experimental value of the critical angle for the silver film falls between the two theoretical values. The accuracy is not great enough to allow a choice between them. The theoretical angle for quartz (the middle arrow) is also larger than the observed. This discrepancy may possibly be caused by a low surface density, or by the composition of the surface being different from that of the interior.

An anomalous behaviour was observed in the case of some of the reflectors when the critical angle had been reached. The reflected line, as observed at the shorter distance, continued to be present for glass and quartz for much greater angles of rotation than for the sputtered elements, although

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[4] J. D. Hanawalt and L. R. Ingersoll, Nature **119**, 234 (1927).

the film of platinum sputtered in hydrogen (curve 2) shows somewhat the same effect. This can hardly be attributed to weak radiation which has a greater spread than 43″ because not all the surfaces show the effect. Neither can it be attributed to lack of planeness in the surfaces since the sputtered films should have all the irregularities of the glass. The effect can be explained if it is assumed that the density over the surface varies and that the residual reflection comes from the more dense areas. A variable surface density is easily understood for glass and quartz, as these were polished The polishing. agent may have become imbedded in them, or the polishing may have altered their surfaces—for instance, to form minute silicon and even oxygen crystals. In the case of platinum any variation of density must be due to sputtering.

Another possible explanation of the phenomenon for glass and quartz is that on account of their composition, the x-rays were able to follow varying types of paths in their short course within the surface—paths ranging from those containing only one kind of atom to those containing only the other. In this way there would arise varying speeds within the reflector and varying indices of refraction, and hence varying critical angles of reflection. No calculations of the extreme types of paths can be made because nothing is known of the nature of the surface which matter presents to x-rays or of the depth of penetration into the surface. However, it is interesting to calculate the critical angles for quartz if the oxygen is assumed to be replaced by silicon (but assuming the number of atoms per cubic centimeter to remain unchanged), and if the silicon is replaced by oxygen. These angles serve as an approximation to the limits within which the variations of path might be expected to affect the critical angle; they are shown in the graph by the arrows to the right and to the left, respectively, of the center arrow. It is seen that the distance between them covers the total fall in intensity. Moreover the value for oxygen is nearest the observed value of the critical angle, as though the surface was covered by oxygen.

In conclusion it may be stated that results have been obtained for sputtered films of nickel and platinum and for quartz which do not agree with theory unless it is assumed that these surfaces have lower densities than the metals and materials themselves. The reflectors of heterogeneous composition, glass and quartz, showed a weak residual reflection beyond the critical angle that must be related in some way to the nature of their surfaces. These phenomena are now being investigated further, with the view of determining the relation between them and the densities of the reflecting surfaces.

This experiment was performed at the University of California. The author wishes to express his sincerest thanks to the members of the Physics Department, particularly to Professor Brackett, for their help and encouragement throughout its entire course.

Brace Laboratory of Physics,
University of Nebraska,
November, 1927.