NOVEMBER, 1928 *PHYSICAL REVIEW* *VOLUME 32*

TOTAL REFLECTION OF X-RAYS FROM NICKEL FILMS OF VARIOUS THICKNESSES

BY HIRAM W. EDWARDS

ABSTRACT

The total reflection of x-rays ($\lambda = 0.707$A) is obtained from mirrors of sputtered nickel films having thicknesses from 0 to 3.3×10^{-5} cm. The measured critical angles were found to vary from a minimum value of 0.0016 radians, that of blank glass, to a maximum value of 0.00339 radians, which was obtained from the thickest nickel film. A satisfactory agreement between the maximum experimental value of the critical angle and that calculated by the Lorentz dispersion formula in which the density of nickel was placed at 8.75 gm/cc, is used as evidence for concluding that the density of the nickel is entirely normal and also that the thickness of sputtered metal films which are to be used in x-ray reflection phenomena must be sufficiently large or misleading results may be obtained. The critical angle from a thick silver sputtered mirror was found to be identical with that obtained from a chemically prepared silver mirror which indicates that the density of silver is independent of the method of deposition.

THE total reflection of x-rays from flat surfaces of glass, quartz and from metals has been investigated by several individuals.[1] From measurements made of the critical angles from these surfaces the index of refraction for x-radiation of differing frequencies has been determined. It has also been shown that the Drude-Lorentz dispersion formula may be used for calculating the index of refraction provided that the frequency of the incident radiation is not close to the critical absorption frequency limits of any of the electrons in the reflecting surface.

In a recent article,[2] Stauss describes his work with sputtered films of nickel, silver and platinum, and gives results from which he concludes that the densities of the nickel and platinum in the sputtered films must have values (4.13 and 5.44 gm/cc for nickel and 14.1 gm/cc for platinum) which are far below their normal values. In partial corroboration with these results he refers to the work of Hanawalt and Ingersoll[3] who report an increase in the lattice constant of nickel in nickel sputtered films, and hence, a decrease in the density. The results of Hanawalt and Ingersoll are not quantitatively in agreement with those of Stauss.

The writer has undertaken to measure the critical angles of several nickel sputtered films which were prepared so that the thickness of the films ranged from a very small value to one of considerably greater value. The amount of metal upon the thinnest film was so little that it was scarcely noticeable on the flat glass surface. The thickest film was entirely opaque to intense sources of visible light.

---

[1] Compton, Phil. Mag. **45**, 1121 (1923); Linnik and Loscharew, Zeits. f. Physik **38**, 659 (1926).

[2] Stauss, Phys. Rev. **31**, 491 (1928).

[3] Hanawalt and Ingersoll, Nature **119**, 234 (1927).

The critical angle for each film was measured by an x-ray reflection apparatus, a description of which appears elsewhere.[4] In this apparatus very long optical paths were provided which reduced the errors in determining the critical angle to values which are less than one percent.

The radiation used was from a water-cooled molybdenum tube which was operated at about 45000 volts. The $K\alpha$ lines were sufficiently intense in comparison with the general radiation to render unnecessary the use of a crystal for providing monochromatic radiation.

The experimental values of the critical angles from the several nickel films together with the one from an unsputtered glass mirror, are tabulated below. The films were all sputtered in air as a residual gas. For the sake of indicating the relative thicknesses of the films the duration of the sputtering time (in minutes) is included. The conditions under which the sputtering took place were maintained as nearly constant as possible, and hence one may assume that the deposit of metal was proportional to this time of sputtering. The thickest of the films was weighed with a probable error of about 10 percent.

TABLE I. *Variation of Critical Angle with Surface Density of Nickel Films.*

Mirror No.	Time of Sputtering (min.)	Critical Angle (radians)	Surface Density (mgm per cm^2)
1	0	0.00160	0.0
2	10	0.00266	0.036
3	15	0.00272	0.054
4	30	0.00308	0.101
5	45	0.00333	0.163
6	80	0.00339	0.290
Calculated value from dispersion theory		0.00340	

Using the Drude-Lorentz dispersion formula, assuming that the density of the nickel was 8.75 gm/cc (which is an average of the values 8.6 and 8.9 given by the Handbook of Chemistry and Physics, published by the Chemical Rubber Publishing Co.) and including the critical $K\alpha$ absorption frequencies for the K electrons, a value of 0.00340 radians was obtained, which is in good agreement with the value obtained by experimentation when using the thickest film. Even if one uses either extreme limit of the values of the density given above the disagreement is not over 1.5 percent. For a density of 8.6 the calculated value of the critical angle is 0.00336 and for 8.9 the value is 0.00344 radians.

These results do not give absolute proof that the density of the nickel in the thinnest film is not the same as it is in the thickest film, but it would seem that a far more reasonable explanation of the smaller values obtained for the critical angles from these films is that the phenomenon of the total reflection of x-rays is not a superficial effect but rather requires a layer of electrons of a somewhat definite thickness. If this is the case, reflection from

---

[4] Edwards, Phys. Rev. **30**, 91 (1927).

the thin films is dependent not only upon some of the electrons of the nickel film, but also upon some of the electrons of the constituent atoms of the glass below the nickel film. The increase of the critical angle with increased thickness of the nickel film supports this hypothesis. The very small change in the critical angle determined by the last two films, even though the film on No. 6 must be nearly twice as thick as that of No. 5, shows that the addition of still more nickel would not measurably change the final result. This statement is substantiated by the agreement of the calculated value with the final experimented value. These results strongly indicate that the actual density of the nickel does not have different values in films of different thicknesses.

The close agreement between the final value of the critical angle and that obtained from the dispersion formula also indicates that actual density of the nickel in the sputtered films used cannot differ by more than 1 percent from the value of 8.75 as used in the theory. Hanawalt and Ingersoll's results indicate an increase in the lattice constant of crystallized nickel in the film of 20 percent and less. Some of their first results indicated that the nickel was not crystalline but amorphous. It is hard to reconcile these two contradictory results.

Some difficulty was encountered in determining the mass of the nickel deposited upon the glass receiving surface. The final method used which gave satisfactory results employed a glass counterpoise which was nearly equal in mass to that of the receiving glass. The glass counterpoise was placed in the sputtering chamber directly under the receiving glass in order to insure a similar treatment, except for the deposit of nickel. The surface density of the nickel on mirror No. 6 was found to be 0.29 mgm per sq. cm.

From the above value of the surface density of nickel, together with a volume density of 8.75 gm/cc the thickness of the nickel film on mirror No. 6 was calculated to be 3.32×10^{-5} cm, which is a little shorter than the shortest wave-length of the visible spectrum. The distance of effective penetration of the x-radiation into the nickel film was found to be 0.00975 cm, assuming that the radiation followed the direction of the line of incidence.

The writer has also measured the critical angle from two different silver films, one of which was deposited upon glass by a chemical method (using the reduction of a silver salt) and the other a sputtered film upon glass. Both films were entirely opaque to ordinary light. The measured critical angles were identical in the two cases and both agreed, within one-half percent, with the value calculated by the dispersion formula. There is undoubtedly no change of density of silver in sputtered silver films.

University of California at Los Angeles,
July 31, 1928.