X-Ray Interference Structure in the Specularly Reflected Radiation from Thin Films

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The phenomenon of x-ray interference upon reflection from thin films is well known. Superposed on this interference a second interference phenomenon has been found which manifests itself as structure in the scattered radiation accompanying the specularly reflected x-ray beam. This new phenomenon is attributed to an interference between the components of scattered radiation originating at the two surfaces of the film. This interference structure in the scattered radiation has been analyzed and used to determine the thickness of vacuum deposited copper films ranging in thickness from 250 to 1000 Å. The results were consistent with those obtained by previously described techniques.

INTRODUCTION

VARIOUS studies of thin films have been made in the past using x-ray interference techniques.[1–3] The procedure has been to reflect a thin sheet of monochromatic x rays from the flat surface of a thin film and measure the ratio of the reflected intensity to the incident intensity. When the glancing angle is in the region of the critical angle for total reflection and the flat film is both sufficiently thin and sufficiently smooth, then x-ray interference can be observed between those fractions of the incident beam reflected at the air-film interface and the film-substrate interface. This x-ray interference is conveniently presented by plotting the normalized reflected intensity as a function of the glancing angle. Examples of such presentations are to be seen in the references cited above and in Fig. 1 of this paper.

The analysis of the results obtained to date has been based on the model of a film which has smooth continuous surfaces and a well-defined complex index of refraction. In spite of its success in predicting the gross features of the reflected intensity as a function of the glancing angle, the model is a manifestly unrealistic one. In view of the fact that the wavelength of the frequently used copper K_α radiation is 1.54 Å, it is unreasonable to expect that the surface is smooth to the order of a wavelength of the incident radiation. If the surface is not ideally smooth, then one might expect that the specularly reflected beam would be broadened and that low-intensity wings should be found on both sides of the geometrically predicted specular reflection. That grossly rough surfaces and films introduce angular spreading of the reflected beam has already been shown.[4] It is the purpose of this paper to examine some of the newly observed features in the low-intensity wings of a sheet of x rays specularly reflected from a thin metallic film which is smooth enough to exhibit the normal x-ray interference structure discussed above. These structural features are explained in terms of the interference between different components of the scattered radiation accompanying the specularly reflected beam.

TECHNIQUE AND RESULTS

The x-ray reflectometer used in the work described below employed radiation from a copper anode x-ray tube. This radiation was variously filtered; a nickel sheet was used to remove the K_β components; an iron

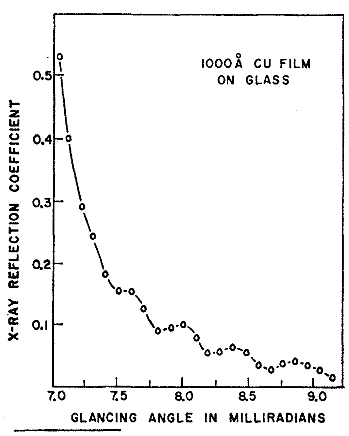

FIG. 1. X-ray reflection coefficient versus incident glancing angle for a 1000-Å Cu film vacuum deposited onto a soft glass substrate. X-ray wavelength = 1.54 Å.

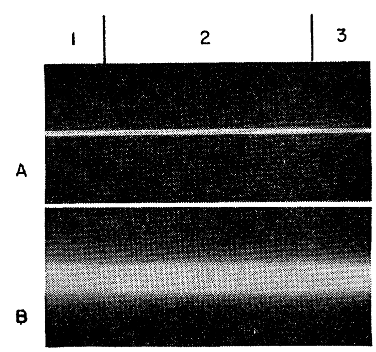

FIG. 2. Photographs of the direct beam for a short and a long exposure taken at a distance of 665 mm from the mirror axis of rotation: (A), 30-sec exposure; (B), 10-min exposure. The photographic film was covered with filter foils as follows: Region 1, 0.001-in. Ni; Region 2, no filter; Region 3, 0.001-in. Fe.

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[2] L. G. Parratt, Phys. Rev. **95**, 359 (1954).
[3] N. Wainfan, N. J. Scott, and L. G. Parratt, J. Appl. Phys. **30**, 1605 (1959).
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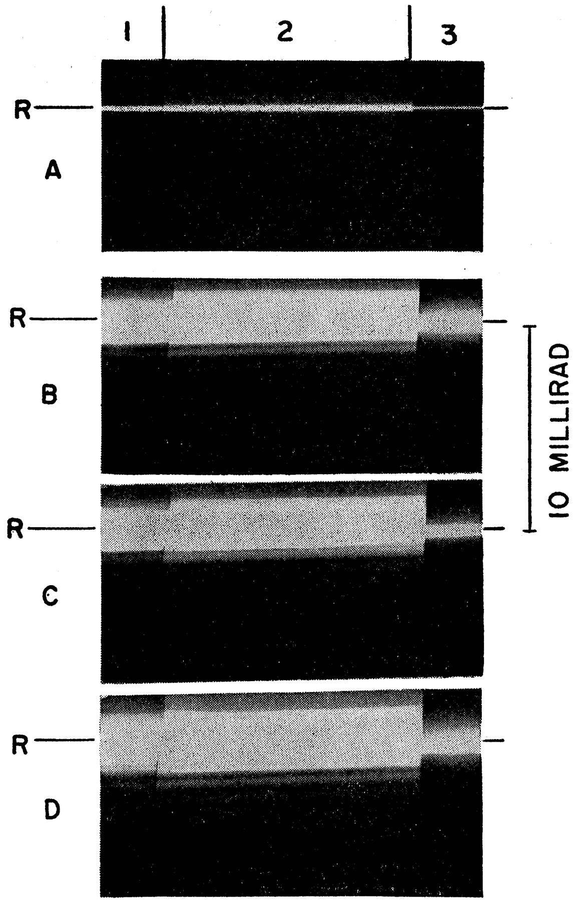


FIG. 3. Photographs of the reflected beam taken at various glancing angles for a 1000-Å Cu film. Sample axis to plate distance is 665 mm, $\lambda=1.54$ Å. (A), 5-min exposure at 8 mrad; (B), 3-h exposure at 8 mrad; (C), 3-h exposure at 8.2 mrad; (D), 4-h exposure at 8.3 mrad. (1) 0.001-in. Ni; (2) no filter; (3) 0.001-in. Fe.

filter was used to remove the soft components including the K; or the radiation was used unfiltered. A tube potential of 40 kV was used for the results presented here. A two slit collimator reduced the x-ray beam to a sheet 0.0025-in. thick having a total angular divergence of 0.2 mrad. The x-ray beam was reflected from the thin-film sample into an ionization chamber or into a stack of photographic plates. The glancing mean angle could be set with a precision of better than 0.025 mrad. For these results the samples were copper films vacuum deposited onto soft-glass substrates. The polished substrates were not of good optical quality and some difficulty was experienced with their lack of flatness. Figure 1 shows the x-ray reflection coefficient plotted against the glancing angle for a typical sample. The interference structure seen in Fig. 1 results from the interaction of the components of the radiation specularly reflected from the two surfaces of the copper film. These are the components labeled R_1 and R_2 in Fig. 5(A). Using the techniques described in the references cited,[1–3] the positions of the interference maxima seen in Fig. 1 yield a film thickness of 1000 Å.

In order to examine the distribution of radiation in the incident and reflected x-ray beams, photographic techniques were used. Figure 2 presents photographs of the incident beam for a short and a long exposure. Figure 3 presents pictures of the reflected beam taken at various glancing angles in the region of the observed reflection interference structure for the copper film presented in Fig. 1. Figure 3(A) is a short exposure of the reflected beam and Figs. 3(B), 3(C), and 3(D) are long exposures taken at various glancing angles. The long exposures reveal the low-intensity radiation distribution around the direction of specular reflection. It should be noted that the angular spacing of the maxima in the low-intensity wings of this radiation distribution, which is on the large-angle side of the main reflected beam, is approximately twice that observed for the maxima

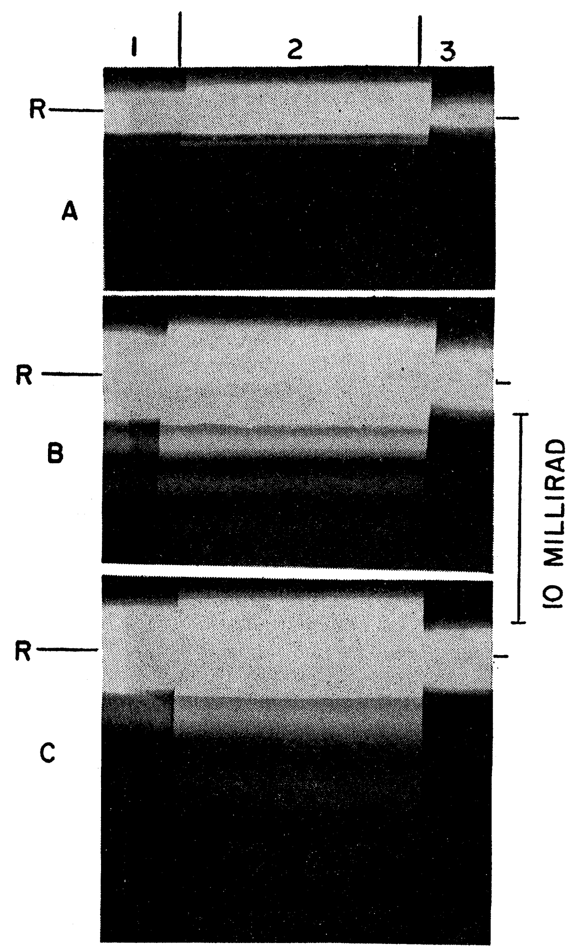


FIG. 4. Photographs of the reflected beam from films of different thickness. Each photograph was taken at a glancing angle corresponding to an interference maximum in the specularly reflected beam. Sample to plate distance was 665 mm and $\lambda=1.54$ Å. (A), 985-Å Cu film on soft glass, 3-h exposure; (B), 398-Å Cu film on soft glass, 3-h exposure; (C), 252-Å Cu film on soft glass, 6-h exposure. (1), 0.001 in. Ni; (2), no filter; (3) 0.001-in. Fe.

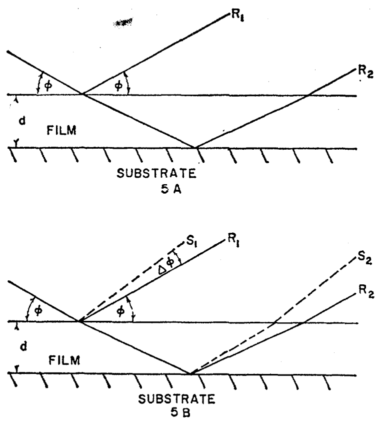


FIG. 5. (A) Diagram of the specular reflection of the incident radiation. R_1 and R_2 are the specularly reflected rays from the air-film and film-substrate interfaces. (B) Diagram of the reflection showing the emergence of scattered radiation, rays S_1 and S_2, from the air-film and film-substrate interfaces.

obtained in the conventional reflection curve such as seen in Fig. 1. Notice is also called to the observation that as the glancing angle is changed the spacing of the maxima in the wings of the radiation distribution remains approximately constant but their "phase" relative to the geometrical position of the reflected beam changes. In Fig. 4 may be seen a group of photographs of the reflected x-ray beam taken for a series of copper films of varying thickness. In each case the glancing angle was set to one of the ordinary reflection maxima. Here it can be seen that as the film thickness increases, the spacing between the maxima in the wings of the radiation distribution decreases.

DISCUSSION

It is assumed that the x-ray interference structure exhibited in the high-angle wings of the radiation distribution around the specularly reflected beam was the result of interference between scattered radiation introduced by the air-copper interface and scattered radiation introduced by the copper-glass interface. This assumption could be checked in several ways. First it is clear from Figs. 2 and 3 that the incident beam has a smaller angular spread than the reflected beams. Second, the angular separation of the interference fringes on the photographs shown in Figs. 3 and 4 are approximately twice those obtained by rocking the sample and measuring the total reflected intensity. That this might be expected can be seen from Fig. 5, since the scattered part of the radiation originating at the copper-glass interface only makes a single traversal through the film while the specularly reflected beam makes a double traversal through the film. Third, the phase relation between the two components of the scattered radiation should depend on the glancing angle of the incident beam. This phase dependence may be seen in Fig. 3. Fourth, calculations of the film thickness using a modified Kiessig approximation for the separation of the interference fringes can be made for the case of the specular reflection and for the case of the scattered radiation. For the specular reflection the order number of the interference maximum is given approximately as

$$N = R + (2d/\lambda)(\phi_m^2 - \phi_c^2)^{1/2},$$

where R is an unknown phase constant, d is the film thickness, λ is the wavelength of the incident radiation, ϕ_m is the angular position of the interference maximum, and ϕ_c is the critical angle. Using a similar approximation the order number of the interference fringes observed in the scattered radiation might be expected to be

$$N' = R' + (d/\lambda)[(\phi_g + \Delta\phi)^2 - \phi_c^2]^{1/2},$$

where R' is an unknown phase constant, ϕ_g is the glancing angle of the incident beam, and $\Delta\phi$ is the angle between the direction of the specular reflection and the observed interference fringe. In these approximations the absorption is neglected.

Film thicknesses were calculated using these relations for a set of films varying between 250 and 1000 Å. These results are shown in Table I. It can be seen that the two techniques agree to within 5%.

TABLE I. Calculations of film thickness in angstroms.

Film	a	b
4 *A*	1030±50	985±50
4 *B*	385±30	398±20
4 *C*	240±25	252±13

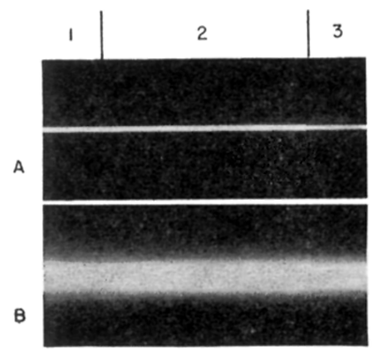
[a] Film thickness obtained for the case of specular reflection.
[b] Film thickness obtained for the case of scattered radiation.

The results obtained indicate a large degree of asymmetry in the scattered radiation around the specularly reflected beam. A proper theoretical analysis of the form factor of this scattered radiation is still to be put forth.

In addition, it might be noted that the precision of the thickness determined by the scattered radiation technique is greater than that yielded by the conventional reflection technique because the fringes have a greater separation and more fringes are generally observable in the new technique than in the one previously reported.

It should be noted from Figs. 3 and 4 that there exists structure in the scattered radiation near the direction of specular reflection other than that subject to analysis in this paper. This structure is under investigation at present.

The observations described here have obvious application in the study of thin films. The method can provide a relatively convenient way of measuring film thicknesses and, in addition, it gives one techniques by which surface-induced scattering might be studied.



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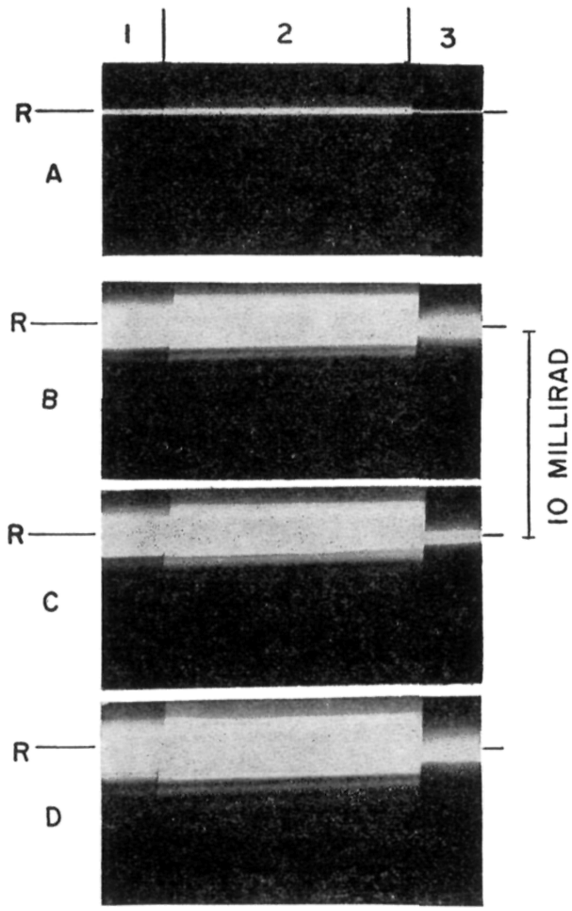

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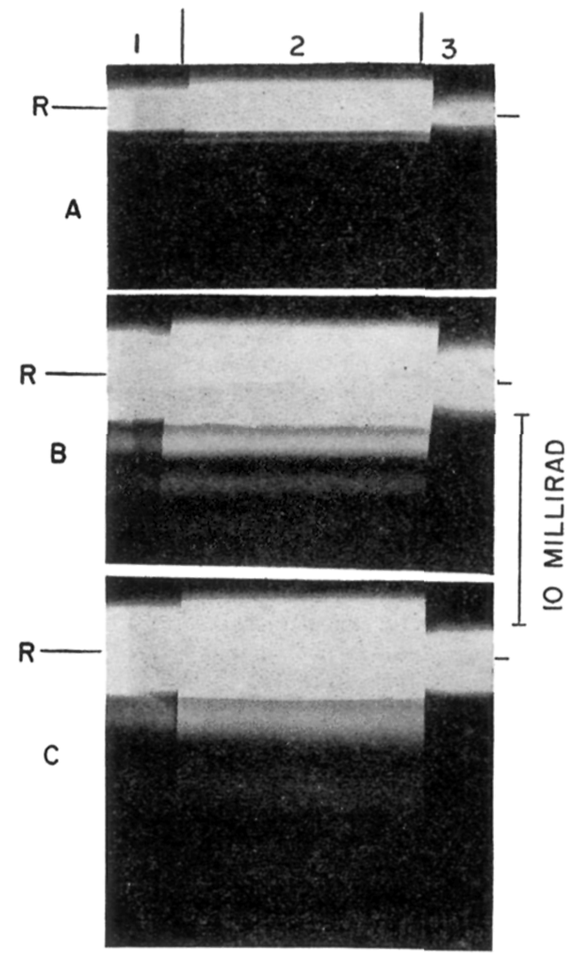

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