Anomalous Surface Reflection of X Rays

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An asymmetric surface reflection of x rays was found. It was experimentally determined that this anomalously reflected wave has almost the same wavelength as the incident wave. The angle of this reflection changes only slightly with the incident angle and the crystal structure of the sample in the measured angular range. The glancing angle of this reflection increases with the wavelength of the incident x rays. According to measurements on evaporated surfaces of metallic elements, these reflection angles were found to have an important relation to the periodic system of the elements.

I. INTRODUCTION

THE equiangular reflection of an electromagnetic wave on the smooth surface of material is well known, but, in the case of x rays, there have been reported[1–8] some results dealing with critical reflections or reflections in materials whose refractive indexes are less than one. Among those papers, there is found no other surface reflection than the equiangular reflection.

However, the existence of an anomalous surface reflection (hereafter abbreviated as A.S.R.) of x rays is reported here. In this case, the glancing angles of the incident x rays with the surface of sample were less than approximately 2°. The chief reasons why this A.S.R. has not been found up to the present are considered to be as follows:

(1) The intensity of the A.S.R. was very low, so that the photographs were, therefore, underexposed.

(2) The inconvenience of the angular range made it difficult to measure the A.S.R.

(3) Samples with strong A.S.R. were not used.

(4) The geometrical accuracy of the apparatus was not satisfactory. More than one of the foregoing reasons are applicable to each previous experiment. In Sec. II of this paper the experimental procedure for the measurement of A.S.R. is described. In Sec. III, the angle and the intensity of the A.S.R. for various samples are presented. Finally, in Sec. IV, the coherency of the A.S.R. is considered and a discussion of the experimental results is given.

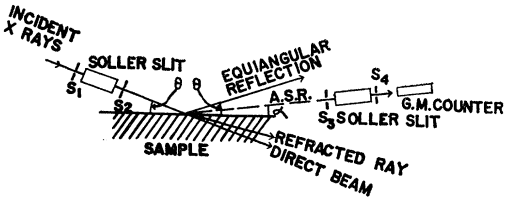

FIG. 1. Schematic view of the experimental arrangement in the incident plane.

II. EXPERIMENTAL PROCEDURE

The essential features of the apparatus used for this experiment are shown in Fig. 1. The x-ray sources used were Ti, Cr, Cu, and Mo radiation. The applied voltages were 20–35 kV and the electron currents were 10–20 mA. For the detection of x rays, a G.M. counter was used and photographic methods were also adopted. The width of slits S_1, S_2, S_3, and S_4 were 0.05, 0.05, 0.05, and 0.1 mm, respectively. The distances x-ray source—S_1, S_1—S_2, S_2—sample, sample—S_3, S_3—S_4, and S_4—window of G.M. counter were 50, 38, 105, 200, 38, and 15 mm, respectively. In the case where photographic methods were used, the above-mentioned distances were enlarged by five times at the most. Since the incident x-ray beam was slender and penetrated to the center of a wide surface of a thick sample, the refracted x rays were too weak to measure. The x rays measured were, therefore, those of equiangular reflection and of A.S.R.

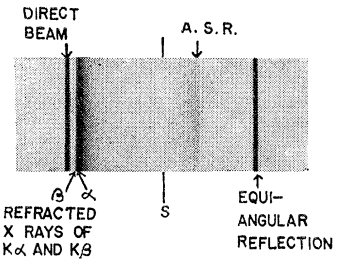

FIG. 2. Photograph of A.S.R. by a glass sample, ×2.

The accuracy of angular measurement by the G.M. counter was estimated to be about 2 min of arc. Commercially available optical prisms made of glass or quartz were used as samples. Also used were single crystals of calcite, LiF, KCl, and NaCl, with cleaved surfaces. For metallic samples the surfaces of metals evaporated on optical prisms were used. The evaporation was done at a pressure 5×10^{-6} Torr using a tungsten boat. The thickness of the metallic films was 200–2000 Å. The size of the reflection plane was about 40 mm long in the direction of the incident x rays and the height was 15 mm.

Most of the metals were measured in air; however,

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Mg, Al, and Ca, which are easily oxidized, were measured in vacuum soon after evaporation. These samples were laid in a bell-jar with two Mylar windows, each 0.05 mm thick. The tungsten boat for the evaporation was placed in the bell-jar. The measurement of reflection could, therefore, be made even during evaporation. This apparatus is so constructed that it can rotate on a goniometer while being evacuated.

III. EXPERIMENTAL RESULTS

(1) With all kinds of samples used, anomalous surface reflections (A.S.R.) were observed. An example of A.S.R. taken by the photographic method with a sample of crown-glass prism is shown in Fig. 2. The line S is the intersection between the extended surface of the sample and the x-ray film. In this case, unlike Fig. 1, the incident x-ray beam was projected at the part nearer to the right side of Fig. 1, indicating clearly the position

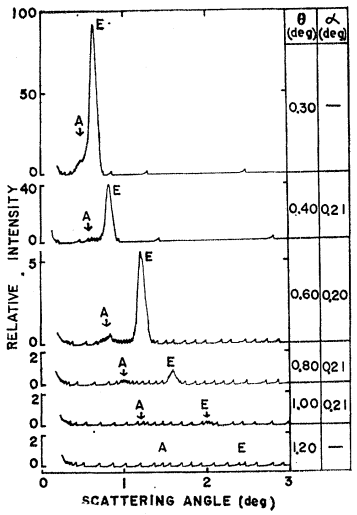

FIG. 3. A.S.R. and equiangular reflection by a quartz sample. X-ray source: Cu radiation, Ni filter, 30 kV, 10 mA.

of the refracted x rays. The distances x-ray source—S_1, S_1—S_2, S_2—sample, and sample—x-ray film were 50, 350, 140, and 700 mm, respectively. The width of slits S_1 and S_2 were 0.1 and 0.05 mm, respectively. The x-ray source was unfiltered Cu radiation, the applied voltage 30 kV, the electron current 12 mA, and the exposure time 72 h. The x-ray film used was Fuji Industrical 200. The glancing angle α measured from this photograph was 0.20°. It was confirmed also by measurement with the G.M. counter.

(2) The angles α and the intensities of the A.S.R. were negligibly affected by the orientation of the crystal samples, within the order of error. This fact is proved more clearly by the similar sharp A.S.R. measured in the case of single crystals and those of evaporated metals which are polycrystalline samples. Even in the measurement with fused quartz, there was hardly any change in α compared with crystalline quartz, though there was an increase of 10% in the intensity of reflection.

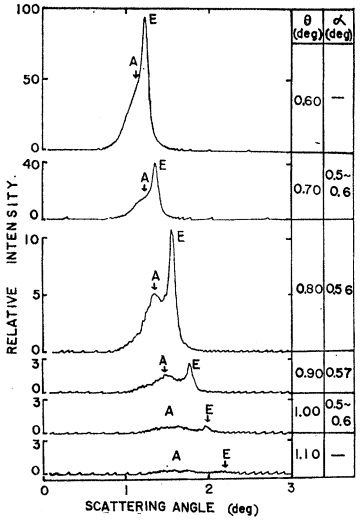

FIG. 4. A.S.R. and equiangular reflection by a Au sample. X-ray source: Mo radiation, no filter, 35 kV, 10 mA.

(3) The effect on the reflections of a change in the incident glancing angle θ is shown in Figs. 3–5, where quartz, Au, and Cr are used as samples. It is seen from the figures that the glancing angle of the equiangular reflection is θ, increasing with increasing θ. On the other hand, α shows little change in the measured angular range. The value of α for each material is, therefore, expressed by almost a single value, without referring to the incident angles. The angles α of quartz, Au, and Cr were 0.21°, 0.56°, and 0.38°, respectively. In these cases the errors of measurements are about $\pm 0.03°$.

(4) The angle α was dependent on the wavelength of the incident x rays, whose effect is shown in Fig. 6. There α is seen to increase with increasing wavelength.

(5) The results of measurements of α on various kinds of metallic elements are shown in Fig. 7. In this case the x-ray source was unfiltered Mo radiation, the applied voltage 35 kV, and the electron current 13 mA. A remarkable characteristic of the curve is that α for

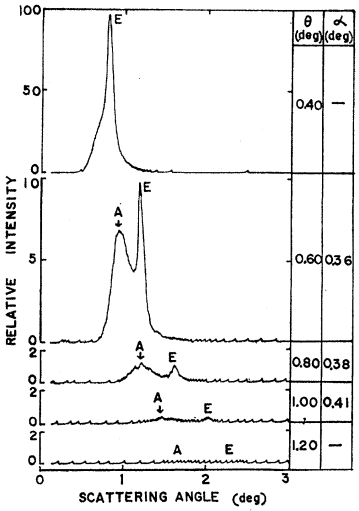

FIG. 5. A.S.R. and equiangular reflection by a Cr sample. X-ray source: Mo radiation, no filter, 35 kV, 13 mA.

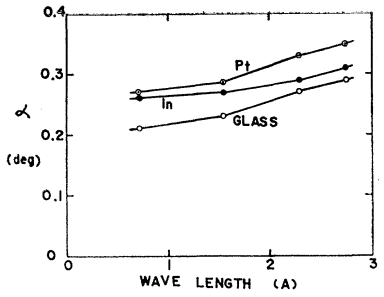

FIG. 6. Dependence of α, the glancing angle of A.S.R., on wavelength. The samples are Pt, In, and glass.

Au, Ag, Cu, and Co is large, constituting the peaks of the curve. When the evaporated metals were used as samples, there was not observed any A.S.R. at the boundary between the metal and the glass. The reason for this is that there is an A.S.R. only from the first surface, and the angle and the intensity of A.S.R. are independent of the material under the sample.

(6) As is seen in Figs. 3–5, the equiangular reflection and A.S.R. have less intensity of reflection when θ is large. When θ is 1.5–2.0°, the majority of the samples have too weak intensities of both reflections to be measured. With the decrease of θ, the intensities of both reflections increase. If θ decreases further, the equiangular reflection enters a critical angle, and it approaches total reflection. On the contrary, the A.S.R. is hard to separate because of its approach to the position of the equiangular reflection. In the case of the glass sample, the angle of its disappearance was roughly estimated as $\theta=0.3°$. For the samples with large α such as Au and Ag, it was about $\theta=0.4°$.

(7) The intensities of the A.S.R. varied according to the kind of material used. Generally speaking, the materials with large α have strong intensities of A.S.R. The intensity variation for most metals is, therefore, similar to Fig. 7 on the whole. However, there are many exceptions. Ir, Pt, Cd, and Cr showed comparatively small intensities of reflection. The thickness of evaporated film had almost no effect on the intensities and angular positions of the A.S.R.

(8) The equiangular reflection was reduced with deterioration of smoothness of the reflection plane by grazing or etching, whereas the A.S.R. was apt to increase temporarily instead of being weakened. These characteristics are shown in Fig. 8 where the glass sample is etched with 3% HF water solution at about 15°C. According to this figure, the intensity of the equiangular reflection decreases rapidly upon etching, while the A.S.R. gradually increases. After the disappearance of the equiangular reflection, the A.S.R. shows a maximum in intensity. If the angle θ is large, the A.S.R. increases in intensity more than the equiangular reflection in shorter etching times. Considering such an effect of surface smoothness upon the intensity of reflection, it is necessary to keep this point in mind in determining the intrinsic intensity of reflection. It is difficult, however, to define the surface smoothness accurately or to measure it relative to the effect of the A.S.R. Accordingly, there is included some ambiguity in the intensity of the A.S.R.

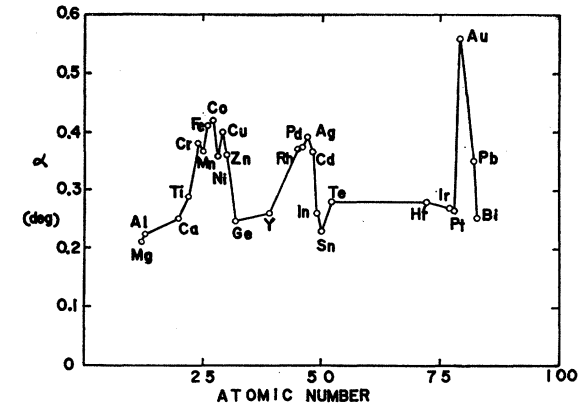

FIG. 7. Dependence of α on the kind of metal.

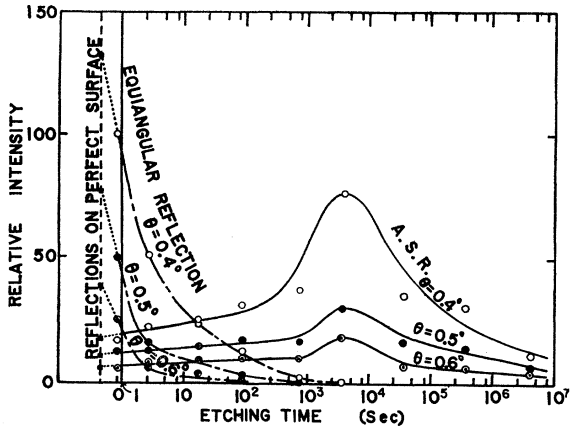

FIG. 8. Semilog plots of the intensities of A.S.R. and the equiangular reflections versus etching time for a glass sample. The dotted lines on the left side are the curves extended toward an almost perfect surface.

(9) On a rough surface there was observed a broadening of the angular width in both reflections.

(10) Wavelength measurements of the A.S.R. were made by crystal reflections. A single crystal of KCl, about 25×25×3 mm in size, was used for the measurements. The x-ray sources used were unfiltered Cr, Cu, and Mo radiations. The sample for A.S.R. was Au which was evaporated on an optical glass prism which had been etched for about 50 min by 3% HF water solution at about 15°C. Under these conditions, since the intensity of the A.S.R. was as strong as that described in experimental results (8), the wavelength measurement was comparatively easy. As to the experimental results, the wavelength of the A.S.R. showed no change within limits of 0.05%.

IV. DISCUSSION

Whether the A.S.R. is or is not coherent with the incident wave is one of the most important problems. Of

course, the ultimate solution of this problem involves the intrinsic mechanisms of A.S.R., which are not known at present. Several reasons for its coherency are, however, as follows:

(i) The wavelength hardly changes.
(ii) The beam is comparatively sharp.
(iii) The glancing angle depends on the wavelength, as expressed in Fig. 6.
(iv) A close relation to the coherent equiangular reflection is found as shown in Fig. 8.

It is seen from Fig. 7 that α has an important relation to the periodic law. This fact leads to a presumption that A.S.R. is different to some extent from the ordinary Thomson scattering. The angles α of the elements Au, Ag, and Cu are large and occupy peak positions of the curve in Fig. 7. From the fact that these three elements all belong to Group I and Series b in the Periodic Table, we can presume that A.S.R. has an important relation to the valence electrons. In this case, the behavior of the valence electrons on the surface of the material has to be considered. No definite explanation seems to be known so far as to what behavior the surplus valence electrons on the surface have, and the measurement of A.S.R. seems to offer a clue for investigating their characteristics.

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Second Sound in Solids

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A critical frequency for thermal fluctuations is calculated above which heat transport proceeds by wave propagation rather than by diffusion. This phenomenon should occur in some dielectric solids. It is the analog of second sound in helium II. A macroscopic point of view is used which relies upon a modification in the Fourier heat equation. Some quantitative results are obtained on the magnitude of this modification.

1. INTRODUCTION

EVER since the initial experiments of Peshkov[1,2] exhibiting second sound in liquid helium, considerable speculation and effort has been devoted to the possible existence of this phenomenon is solids. This effort has been stimulated, in the main, by those theories[3–6] of second sound which depend only upon the presence of a phonon gas. Since phonon gas excitations exist in any solid, second sound should be detectable in solids as well as in liquid helium. We propose that thermal waves should, indeed, be detectable in many substances but that there is a critical frequency for the onset of the phenomenon. This idea is current and has been elegantly presented by Prohofsky[7] from a microscopic point of view. Here we wish to relate the idea, heuristically, to a well-known paradox of heat transport and to derive some simple quantitative predictions on this basis. Our point of view is essentially macroscopic.

2. THERMAL DIFFUSION

It is well known that the diffusion equation for thermal transport results from the following two elementary considerations: The first is a statement of the continuity equation for heat transport in the absence of density or pressure gradients. This is given by

$$C\frac{\partial T}{\partial t}+\nabla\cdot\mathbf{q}=0, \tag{1}$$

where C is the heat capacity per unit volume, T is the absolute temperature, $\mathbf{q}$ is the thermal heat current density, t represents time, and ∇ is the spatial gradient operator. If this equation is combined with the phenomenological Fourier equation of thermal conductivity for heat flow in a solid,

$$\mathbf{q}=-K\nabla T, \tag{2}$$

the diffusion equation results. In Eq. (2), K is the thermal conductivity. But a much more physically

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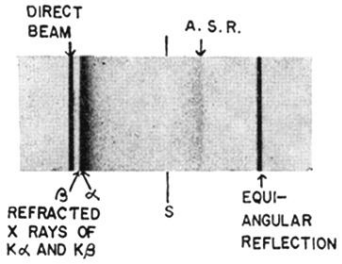

FIG. 2. Photograph of A.S.R. by a glass sample, ×2.