energy loss problem for helpful remarks. He is indebted especially to Prof. Fujimoto of University of Tokyo for his valuable suggestions.

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DISCUSSION

L. MARTON: How do you measure the film thickness and what is the accuracy of your thickness measurements?

H. WATNANABE: The film thickness was measured with a multiple-beam interferometer. The accuracy for the thinnest film is approximately 10%, and that for thicker one is a few per cent.

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Anomalies in Kikuchi Reflection Diagrams
I. Intensity Anomalies*

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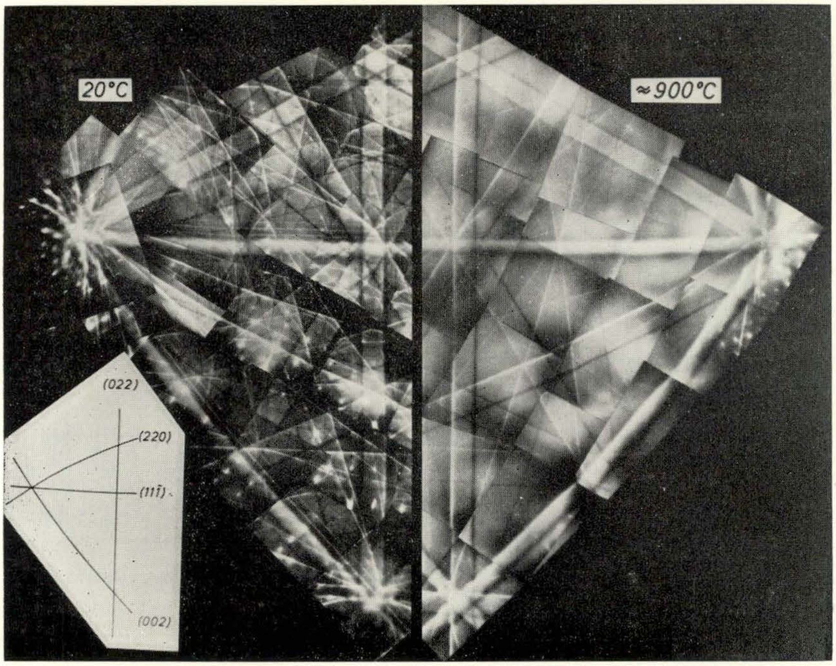
The intensity of Kikuchi lines is decreased or increased in an angular region up to 2 degrees, if the ends of these anomalous regions are directions of simultaneous reflections. This phenomenon was investigated systematically on copper single crystal spheres having untouched surfaces at room and elevated temperatures.

Investigations were carried out on spherically-shaped single metal crystals of about 5 mm diameter, which were prepared by asymmetrical cooling of a drop of molten metal on a ribbon of tungeten or carbon in high vacuum. Since the surface is subjected to neither mechanical nor chemical treatments, it has high crystalline quality and is free from contamination. The spherical shape permits us to study all crystallographic planes as surface. Fig. la is composed of about 20 single pictures and shows a characteristic triangle in the Kikuchi pattern from a copper sphere at 20°C (60 kev). All possible intersections of bands and lines may be studied in this triangle. The tungsten or carbon carrier can be heated by an electric current while the crystal is under observation. Fig. 1b is also a composed picture, which corresponds to the mirror image of Fig. la across (022), taken at about 900°C. The Kikuchi-lines of higher indices are relatively less pronounced than those of lower indices. The diagram becomes simpler without losing sharpness. Both advantages, the spherical shape and the observation at elevated temperature, were utilized in the systematic investigation of the appearance of intensity anomalies in the Kikuchi reflection-diagram.

By intensity anomalies, we mean the decrease or increase in intensity of reflection lines in a large range of angle up to 2°. Fig. 2a~d show four examples of decrease, Fig. 2e one of increase of Kikuchi-lines in a copper reflection diagram. This phenomenon of decrease was first observed in transmission with a convergent electron beam from PbI_2[2), and then studied together with the increase in greater detail on mica and explained as the effect of simultaneous reflection[3)]. This was later confirmed for PbI_2 and interpreted in terms of the dispersion surface[4)]. In reflec-

* Read by E. Menzel.



(a) (b)

Fig. 1. Kikuchi-pattern of a copper sphere in the characteristic triangle. a) 20°C, b) about 900°C.

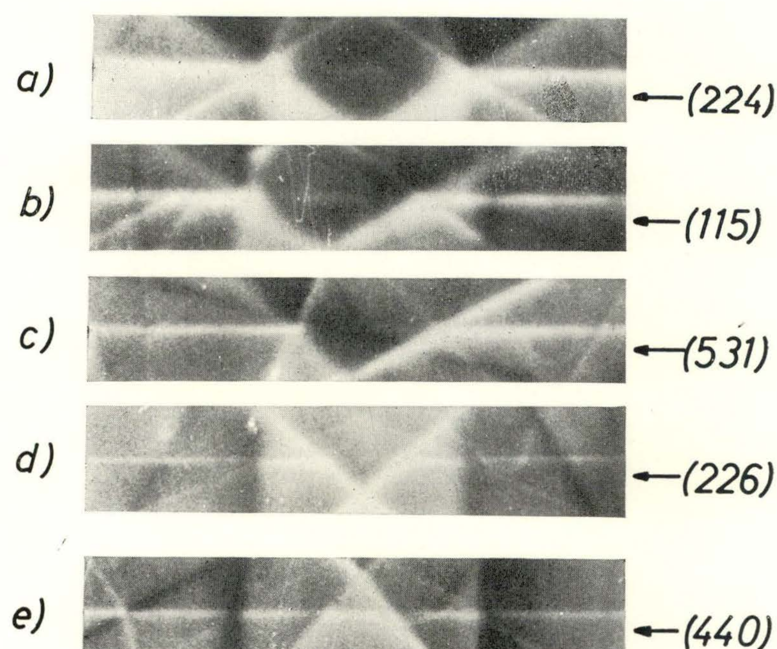
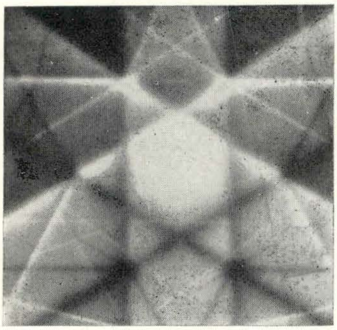


Fig. 2. Types of intensity anomalies. a–d) decrease e) increase.



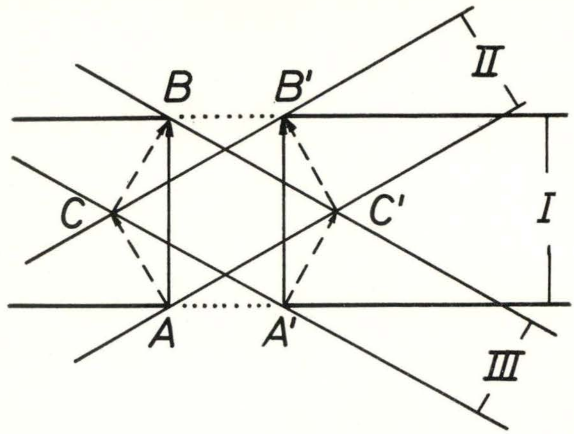
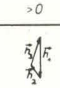
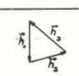


Fig. 3. Direct and simultaneous reflections revealing the decrease of intensity.

tion case this phenomenon was first described for Si[5]. For transmission the calculations are carried out on the basis of the dynamical theory[6] and of the second Born approximation[7]. In both cases the results agree qualitatively with the experiments.

The Kikuchi-diagram in Fig. 3 shows the surroundings of the (224)-line of Fig. 2a. For transmission we found in 1951 the following rules[3] (see schematic diagram in Fig. 3). The limits of the anomalous range AA′ and BB′ are given by the fact that for the reflection in question both direct reflection (AB and A′B′) and simultaneous reflection with one or more coupling reflections (AC-CB and A′C′-C′B′) are possible. The perturbing reflections are the same for the two limiting points but the sequence is reversed. If the anomalous range lies outside the two perturbing bands (here "band" designates the region between a pair of Kikuchi-lines), then the reflection intensity is decreased in the anomalous region; if it lies within the bands, it is increased. These rules are represented again formally in Table I, where the $\boldsymbol{h}_i$'s represent lattice vectors in reciprocal lattice.

Table I.

	decrease	increase
$\sum_{i=1}^{3} \vec{h}_i$	0	0
$\measuredangle(\vec{h}_1\vec{h}_2)$	$>\pi/2$	$>\pi/2$
$\measuredangle(\vec{h}_1\vec{h}_3)$	$>\pi/2$	$>\pi/2$
$\measuredangle(\vec{h}_2\vec{h}_3)$	$<\pi/2$	$>\pi/2$
cub. system		
$\Sigma\, h_1 \cdot h_2$	<0	<0
$\Sigma\, h_1 h_3$	<0	<0
$\Sigma\, h_2 h_3$	>0	<0
vectors in the reciprocal lattice		

The appearance of the anomalies and the validity of the rules were tested on copper single crystals, which had been grown on carbon substrates. Table II lists some intensity anomalies observed for Cu in reflection. Altogether, 22 crystallographically dif-

Table II.

Decrease

Nr.	$h_1k_1l_1$	$h_2k_2l_2$	$h_3k_3l_3$	Zone
1.	242	$\bar{2}\bar{2}0$	$0\bar{2}\bar{2}$	$1\bar{1}1$
2.	$53\bar{1}$	$\bar{2}02$	$\bar{3}\bar{3}\bar{1}$	$3\bar{4}3$
3.	$\bar{2}26$	$\bar{1}\bar{3}\bar{3}$	$31\bar{3}$	$3\bar{3}2$
4.	244	$\bar{3}\bar{3}\bar{1}$	$1\bar{1}\bar{3}$	$4\bar{5}3$
5.	244	$\bar{2}\bar{2}0$	$0\bar{2}\bar{4}$	$2\bar{2}1$
6.	$53\bar{3}$	$\bar{3}\bar{3}\bar{1}$	$\bar{2}04$	$6\bar{7}3$
7.	620	$\bar{4}\bar{2}\bar{2}$	$\bar{2}02$	$1\bar{3}1$
8.	622	$\bar{5}\bar{1}1$	$\bar{1}\bar{1}\bar{3}$	$1\bar{4}1$
9.	424	$\bar{3}\bar{1}\bar{1}$	$\bar{1}\bar{1}\bar{3}$	$1\bar{4}1$
10.	$51\bar{1}$	$\bar{2}02$	$\bar{3}\bar{1}\bar{1}$	$1\bar{4}1$

Increase

Nr.	$h_1k_1l_1$	$h_2k_2l_2$	$h_3k_3l_3$	Zone
1.	331	$\bar{1}\bar{3}\bar{3}$	$\bar{2}02$	$3\bar{4}3$
2.	$\bar{3}15$	$1\bar{3}\bar{3}$	$42\bar{2}$	$6\bar{7}5$
3.	440	$\bar{1}\bar{3}\bar{3}$	$\bar{3}\bar{1}3$	$3\bar{3}2$
4.	$42\bar{2}$	$\bar{1}13$	$\bar{3}\bar{3}\bar{1}$	$4\bar{5}3$
5.	$\bar{2}04$	$0\bar{2}\bar{4}$	220	$2\bar{2}1$
6.	222	$\bar{3}\bar{1}3$	$1\bar{1}\bar{5}$	$2\bar{3}1$
7.	224	$\bar{4}\bar{2}\bar{2}$	$20\bar{2}$	$1\bar{3}1$
8.	424	$1\bar{1}\bar{5}$	$\bar{5}\bar{1}1$	$1\bar{4}1$
9.	115	$\bar{3}\bar{1}\bar{1}$	$20\bar{4}$	$2\bar{7}1$
10.	$31\bar{3}$	113	$\bar{4}\bar{2}0$	$3\bar{6}1$

ferent decreases of intensity and 10 increases were observed. They all follow the rules. Fig. 4 shows the curve of the intensity variation of the (115)-reflection decreased by the simultaneous reflections ($1\bar{1}\bar{3}$) and ($\bar{2}0\bar{2}$). I_2 is the intensity at the Kikuchi line, I is the intensity of the background, interpolated from the intensities on both sides of the Kikuchi line. Each point in Fig. 4 represents a photometric recording carried out perpendicular to the Kikuchi line. For the region of dotted curve, determination of I is relatively inaccurate because of the strong crossing lines and the resulting strong intensity gradients. In Fig. 4a the temperature of the crystal is 150°C, and crossing lines produce the large fluctuation; in Fig. 4b the temperature of the same crystal is 700°C and the intensity of the main reflection is reduced, but the relative fluctuation is reduced still more.

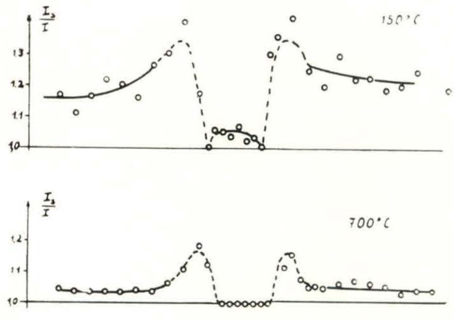


Fig. 4. Intensity variation of a (115)-line, decreased by ($1\bar{1}\bar{3}$) and ($\bar{2}0\bar{2}$).

The converse question was also examined: Is an intensity anomaly always observed if the above rules are fulfilled? Since in a face-centered cubic crystal the sum of two reflections always yields again a permitted reflection, there must be an anomaly for every intersection of two Kikuchi lines. We found the decrease of intensity in all cases expected from the rule (investigations were carried out only on those intersections in which the sum of the squares of Miller indices of the intersecting lines was less than 21). The decrease is generally more pronounced than the increase. The increase could not always be observed for all expected cases, such as $(\bar{2}04)+(331)+(\bar{1}\bar{3}\bar{5})=0$. The rules applied, however, to all observed cases.

We found also the cases in which the indirect path consists of several reflections (see also reference 5), e.g. the increase of ($40\bar{4}$), for which the indirect path consists of three reflections (($\bar{3}\bar{1}\bar{1}$) $+(113)+(\bar{2}02)$). Here, too, the region of increase lies within two perturbing bands.

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DISCUSSION

S. Miyake: Several years ago, Dr. Kambe, who is now in Berlin, and myself studied the influence of the phase factors of Fourier potential on electron diffraction pattern in which two Bragg reflections take place simultaneously. Kambe applied the same theory to discuss the structure of the intensity distribution at a cross of two Kikuchi lines, and I suppose that his result, which is not published yet, will be useful for explaining the intensity-structure of Kikuchi lines at their crosses.

E. Menzel: Thank you for your advice. The phase change at a cross of the Kikuchi lines is also important for the change of intensity along a larger part of the lines. This was shown by a mechanical model of three coupled pendulum.