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Fine Structure of Electron Diffraction Beams from a Gold Crystal and from a Silver Film on a Gold Crystal

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A study of the diffraction of low-speed electrons from a gold crystal has been made under the same conditions as those previously used for a silver crystal to determine the extent to which the previously observed fine structure characteristics depend on the nature of the atoms composing the crystal. For normal incidence there are many differences in the positions, structure, and relative intensities of the corresponding beams from the two crystals. *Intensity measurements as a function of angle of incidence* show some similarities in these characteristics for the two crystals, but there are many more differences. Results for a thin silver film on a gold crystal are in general the same as those for a massive silver crystal so that the different results for the two crystals appear to be real and not due to differences in surface irregularities. Differences are noted in the deviations from the surface grating formula during growth and decay of diffraction beams from the gold crystal and from the silver film on the gold crystal. This makes it appear unlikely that the number of atom lines in the elementary grating is the only determining factor. Beams which deviate from the surface grating formula may follow one of two depth grating formulas. Some dependence of this characteristic on angle of incidence is noted. Since no beams characteristic of a surface gas lattice on the gold crystal were observed when gas was known to be present, the gas layer must be noncrystalline.

Introduction

EXPERIMENTAL results reported in a recent paper[1] showed that for a silver crystal the number of experimental maxima in the low-voltage range is considerably greater than that to be expected from simple theory; and further that these additional maxima may, in general, be grouped as components of fine structure of the main diffraction beams. It was pointed out that a comparison of results for silver and gold crystals should be of value in determining the cause of this fine structure since the lattice constants of these crystals are the same to within 0.4 percent and both lattices are face-centered cubic. Thus a possible dependence of the fine structure on the lattice constant of the crystal should not be responsible for any appreciable differences in the characteristics of the corresponding diffraction beams from the two crystals, and hence any remaining differences must result from the different types of atoms.

The apparatus and method of procedure are the same as that described in detail in the former paper. In the investigations of a silver crystal two different experimental procedures were used. In one of them the characteristics of the diffraction beams were obtained as a function of primary voltage and angle of incidence which varied from zero (measured from the normal) to several degrees either side of the normal. The diffraction beams correspond to reflections from various sets of atomic planes in the crystal *not* including the one parallel to the crystal face. The other procedure was that of regular reflection from the set of planes parallel to the crystal face and with angles of incidence other than normal. This is the same as that used by Davisson and Germer[2] when they observed what they called "anomalous dispersion." In the present experiments only the first procedure was used since it is possible with the results of this method to associate the fine structure characteristics with the individual diffraction beams with greater certainty.

Experiments with a Gold Crystal

Preparation of crystal

The crystal was made from proof gold of purity 0.99995 obtained from the U. S. Mint. The method was the same as that used for making copper and silver crystals.[1] The dimensions and

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[1] H. E. Farnsworth, Phys. Rev. **40**, 684 (1932).

[2] C. Davisson and L. H. Germer, Proc. Nat. Acad. **14**, 619 (1928).

TABLE I. *Diffraction beams for normal incidence.*

		Silver Lattice				Gold Lattice			
Beam and order numbers*	Plane of reflection	Experimental voltage	Theoretical voltage	Voltage difference	Uncorrected relative intensity	Experimental voltage	Theoretical voltage $V=150/\lambda^2$	Voltage difference	Uncorrected relative intensity
			(100) Azimuth				*(100) Azimuth*		
1–1	(210)	46.1	56.3	10.2	100	58.0	56.8	−1.2	100
1–1	(210)	55			78				
2–1	(310)	75.7			3.5				
2–1	(310)	93.5	100.2	6.7	5	101.5	100.9	−0.6	23
3–1	(410)	141.0	162.8	21.8	21	136.0	164.0	28.0	17
4–1	(510)	229.5	243.8	14.3	7	247.0	245.6	−1.4	21
5–1	(610)	335.5	343.0	7.5	6				
1–2	(320)	152.5	169.3	16.8	4	171.0	170.5	−0.5	4
2–2	2(210)	206.5	225.4	18.9	19	191.0	227.1	36.1	9
3–2	(520)	280.5	303.3	22.8	7	303.0	305.5	2.5	11
			(111) Azimuth				*(111) Azimuth*		
1–1	(311)	20			22				
1–1	(311)	27			35	27.5			73
1–1	(311)	34.0	30.3	−3.7	70	32.0	30.5	−1.5	86
2–1	(511)	56.0	65.7	9.7	78	55.0			39
2–1	(511)	62.0			70	64.0	66.2	2.2	48
3–1	(711)	115.0	119.6	4.6	16	117.0	120.5	3.5	22
4–1	(911)	168.3	191.7	23.4	4				
4–1	(911)	175.5			3.5				
1–2	(211)					70.0			26
1–2	(211)	77.5	81.1	3.6	9	78.5	81.7	3.2	29
2–2	2(311)	112.5	121.2	8.7	16	120.0	122.1	2.1	8
3–2	(411)	158.5	182.6	24.1	9	163.0	183.9	20.9	15
3–2	(411)	173.0			6				
4–2	2(511)	250.5	262.9	12.4	9	256.0	264.8	8.8	11

* The notation is the same as that in Table I of former article.[1]

mounting of the single crystal piece used in the experimental tube were also the same as those for the previous experiments with copper and silver. The surface to be studied was cut parallel to a (100) set of planes, and mounted so that it could be rotated about an axis perpendicular to the (100) azimuth as in the case of one of the silver crystals. A smooth etched surface parallel to the (100) set of planes is much more difficult to obtain on a gold crystal than on silver. The face was etched in potassium cyanide dissolved in hydrogen peroxide. The solution was renewed every few minutes until the desired etching was obtained. *Aqua regia* was found to etch only parallel to (111) planes.

Diffraction beams for normal incidence

Table I shows the positions and relative intensities of the beams for normal incidence. The corresponding results for a silver crystal are included for comparison. When there are more than one component they are listed separately, but the differences between theoretical and experimental voltages are given for only the strongest component of each beam. There is no close correspondence in the relative intensities of the corresponding diffraction beams from the two crystals.

Curves showing the growth and decay of some first and second order beams in the (100) azimuth of a gold crystal are shown in the upper part of Fig. 1. These curves were obtained by varying the primary voltage in small steps, and measuring the current to the Faraday collector, whose angular position was adjusted for the maximum of the beam at *each* reading, while the total primary current was held *constant* for the entire set of observations. The corresponding curves for a silver crystal are shown directly beneath those for gold.

From these results it is clear that there are many differences in the positions, structure and relative intensities of the corresponding beams from the two crystals for normal incidence.

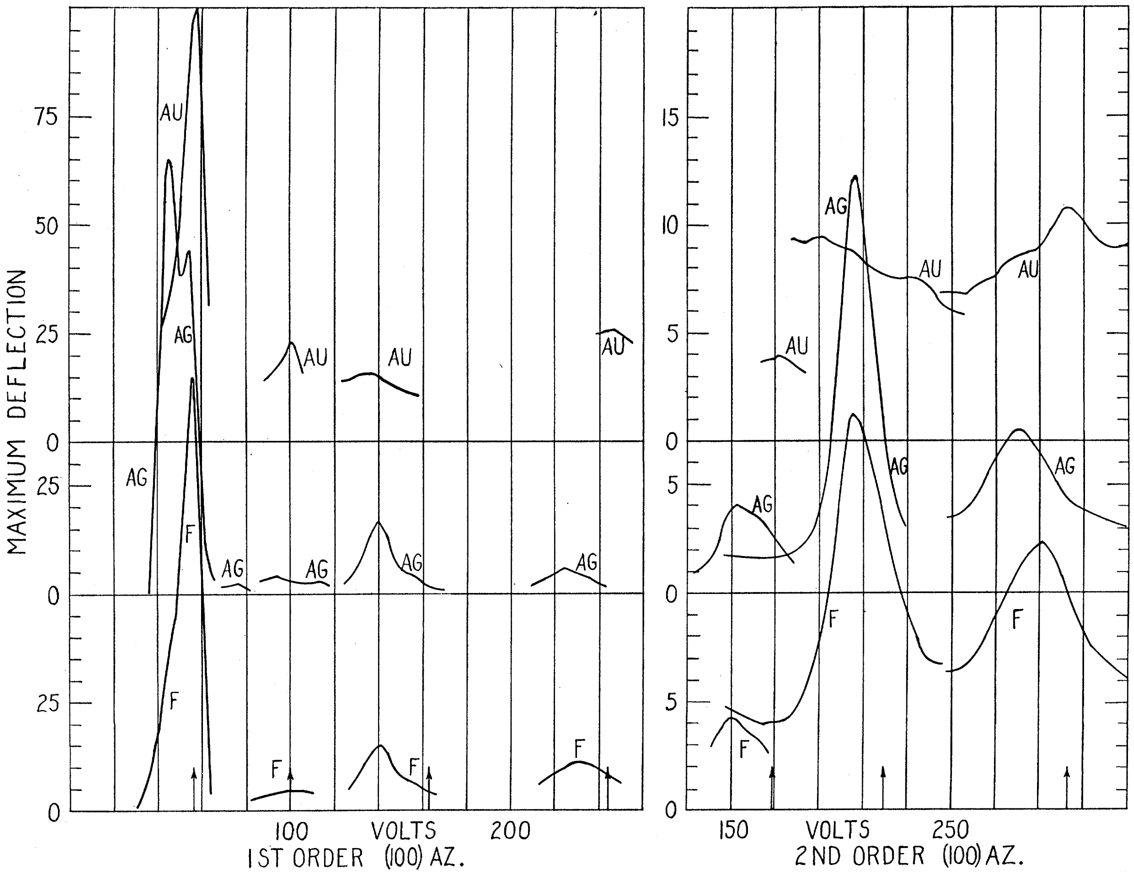

FIG. 1. Curves showing maximum deflection as a function of primary voltage for a gold crystal (Au), a silver crystal (Ag), and a silver film on a gold crystal (F). The colatitude angle is adjusted for maximum deflection for all points of the curves. Arrows indicate positions of theoretical beams for refractive index of unity.

Gas beams for gold

A search was made for beams due to a surface gas lattice at positions corresponding to half-order reflections but none was found even when the crystal surface was known to be covered with a considerable layer of gas, the presence of which results in a decrease of the intensity of the diffraction beams characteristic of the gold lattice. Thus gas does not always form in a simple lattice on the surface of a single crystal as it does in the cases of copper, silver and nickel.

Intensity measurements as a function of angle of incidence

Fig. 2 shows the relative intensities of various beam components as a function of angle of incidence. These curves were obtained by the same method as that previously used for silver crystals[1] and are to be compared with the curves for the corresponding beams from a silver crystal reproduced in Fig. 3 for comparison. This comparison of the curves for the corresponding beams from the gold and silver crystals shows both similarities and differences. Taking the 1st beam, 1st order, corresponding to reflection from the (210) set of planes, we note that there are four components in each case with considerable similarity in their distribution in angle of incidence. However, the voltages of the strongest component in the two cases are noticeably different. For the 2nd beam, 1st order, we also note the similarity that both of the strongest components increase rapidly as the angle of incidence varies from zero to negative values. However, many more weaker components of this beam were observed for silver than for gold. In

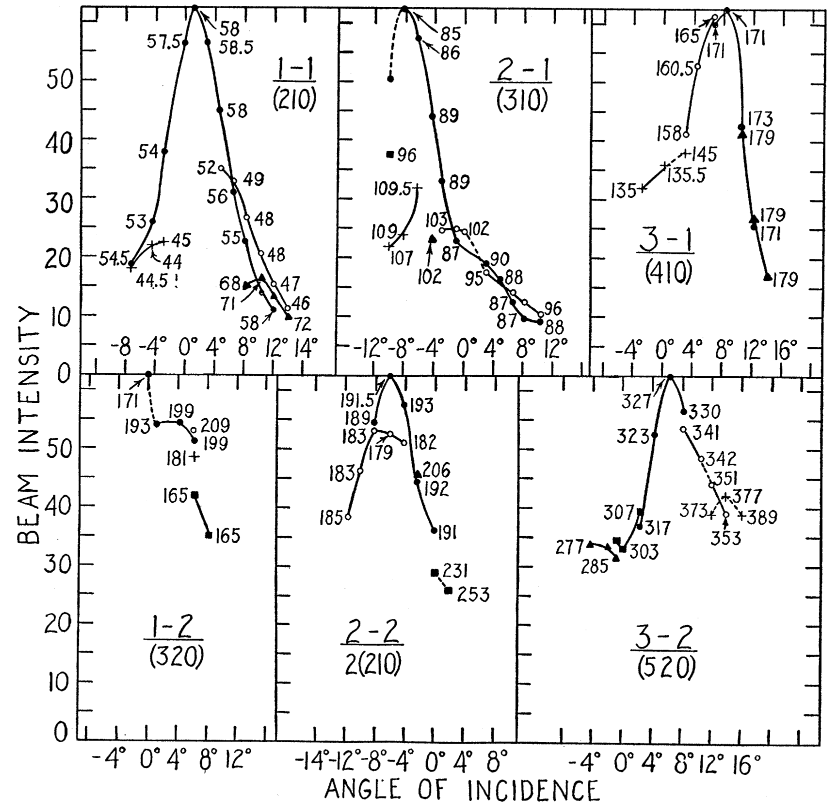

FIG. 2. Results for a gold crystal. The curves show relative intensities of beam components as a function of angle of incidence for the (100) azimuth (uncorrected for background scattering). Plane of reflection is in the (100) azimuth. Designation of beam 3–2 signifies third beam in the second order as per notation in Table I. Values of corrected voltages are given for several of the points.

the case of the 3rd beam, 1st order, there appears to be no correspondence for the two crystals. The number of components for the silver crystal is much the greater and the few components for the gold crystal do not closely resemble any of those for silver. Comparisons of three beams in the second order show no close resemblance. In each case the number, position, and form of the components are different.

EXPERIMENTS WITH A SILVER FILM ON A GOLD CRYSTAL

Although there are some similarities in the above characteristics for gold and silver crystals, the results show many differences. In comparing such characteristics for crystals of different substances it is essential to eliminate possible surface effects which may introduce differences apparently characteristic of the substance composing the crystal. By considering various surface irregularities it is possible to account for certain additional maxima, as recently discussed by Rupp.[3] The etched surface of the gold crystal was known to be much more irregular than that of the silver crystal, and it is natural to suppose that the nature of the irregularities, the size of the elementary lattices, and the mosaic structure of the two crystals may be quite different. However, a thin film of silver deposited on the surface of a gold crystal may be expected to possess the same irregularities and roughness as the underlying crystal, and consequently the corresponding results for such a silver film should furnish the desired information.

To carry out such measurements, the experimental tube was so altered that silver could be deposited on the gold crystal by evaporation in a

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[3] E. Rupp, Ann. d. Physik 13, 101 (1932).

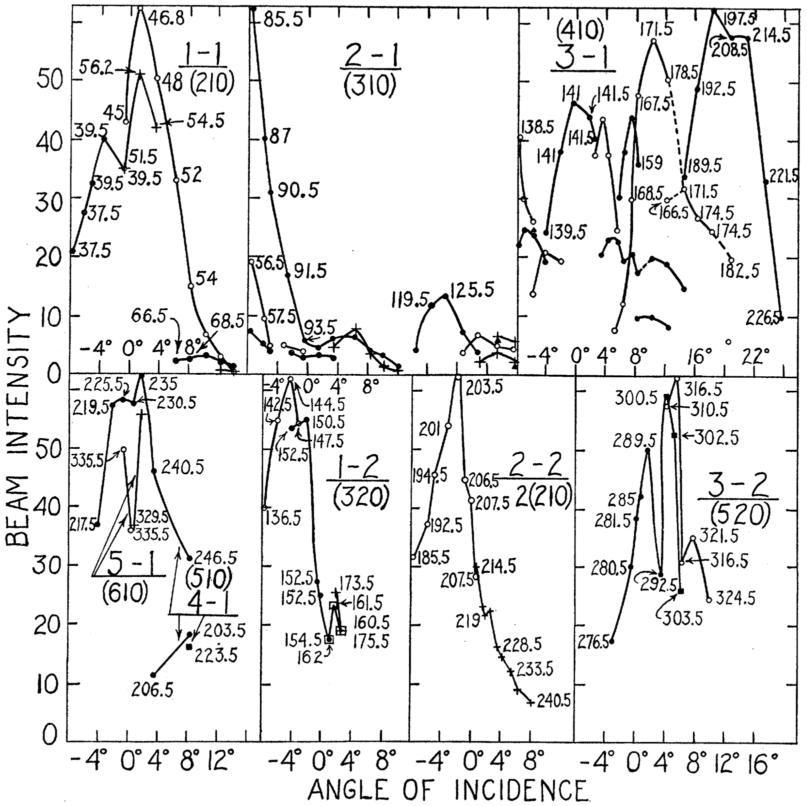

FIG. 3. Results for a silver crystal corresponding to those for a gold crystal shown in Fig. 2 with the same notation.

high vacuum and measurements were taken on the film immediately following the deposit. This was accomplished by withdrawing the gold crystal into a side tube where the front face was exposed to silver vapor coming from a pure silver mass held at dull red heat by electron bombardment from the rear. The usual order of procedure was to outgas the silver mass at red heat with the gold crystal concealed. The gold crystal was then outgassed at red heat, after which it was placed four or five cm from the dull red silver for from 1 to 10 min. The process was repeated several times by evaporating off the film on the gold crystal and then redepositing, with the same experimental results. The silver film forms in a crystalline structure whose orientation is determined by that of the underlying gold crystal. The film thicknesses were not known but were too thin to be visible.

The lower curves in Fig. 1 show the results obtained in the (100) azimuth for a silver film on a gold crystal when using normal incidence. As for the case of the other curves in the figure, we are plotting the maximum deflection of the beams for various voltages, the collector angle being adjusted for each reading. The arrows indicate the theoretical positions of the beams. The first four beams of the 1st order, and the first three beams of the 2nd order are shown. The relative intensities and positions of the beams are quite different for the silver and gold crystals. It is seen that the results for a silver film are quite closely in accord with those for a silver crystal, the chief difference being in the 1st beam, 1st order. For silver, this beam has two components, the low voltage one being more intense, while for the silver film there is an indication of a second component with the higher voltage one being much more intense. The voltage for this component is also approximately the same as that for the corresponding beam from the gold crystal. There is also some difference in the 3rd beams of the 2nd order, but in general it is clear that the results for the film are very nearly in accord with those for a massive silver crystal. Observations on the film were also taken for various angles of incidence and curves plotted similar to those in Figs. 2 and 3 for several beams. These curves are not shown but were found to be closely in accord with those for the silver crystal. In particular, the complicated structure of the 3rd beam, 1st order, for the silver crystal was accurately reproduced for the silver film. Hence the differences between the results for silver and gold crystals appear to be real and not due to differences in irregularities of a secondary nature. Further results are required to determine the cause for the above-mentioned differences in the results for the film and for the silver crystal. In particular, measurements on thinner films of known thickness should be of value.

Deviations from the surface grating formula

It was first observed by Davisson and Germer that, during the growth and decay of a particular diffraction beam, the variation of the colatitude angle with the primary voltage was not in general such as to keep $V^{\frac{1}{2}} \sin \phi$ constant as is required by the plane grating formula. This condition was found to obtain best for the lowest voltage beams and not at all for higher voltage beams. This was interpreted to mean that the diffracting system was not a single lattice, but a collection of many small lattices of not more than five or ten atom lines in width, similarly oriented, but otherwise unrelated.

Fig. 4 shows a plot of $\sin \phi$ against $(150/V)^{\frac{1}{2}}$. The straight lines are surface grating lines given by $n_1\lambda = (a/2) \sin \phi$, and the curved lines are depth grating lines given by $n_2\lambda = a/2 + a/2^{\frac{1}{2}} \cos (\phi + 45°)$, and $n_3\lambda = a/2(1 + \cos \phi)$. The rows of atoms corresponding to the various grating

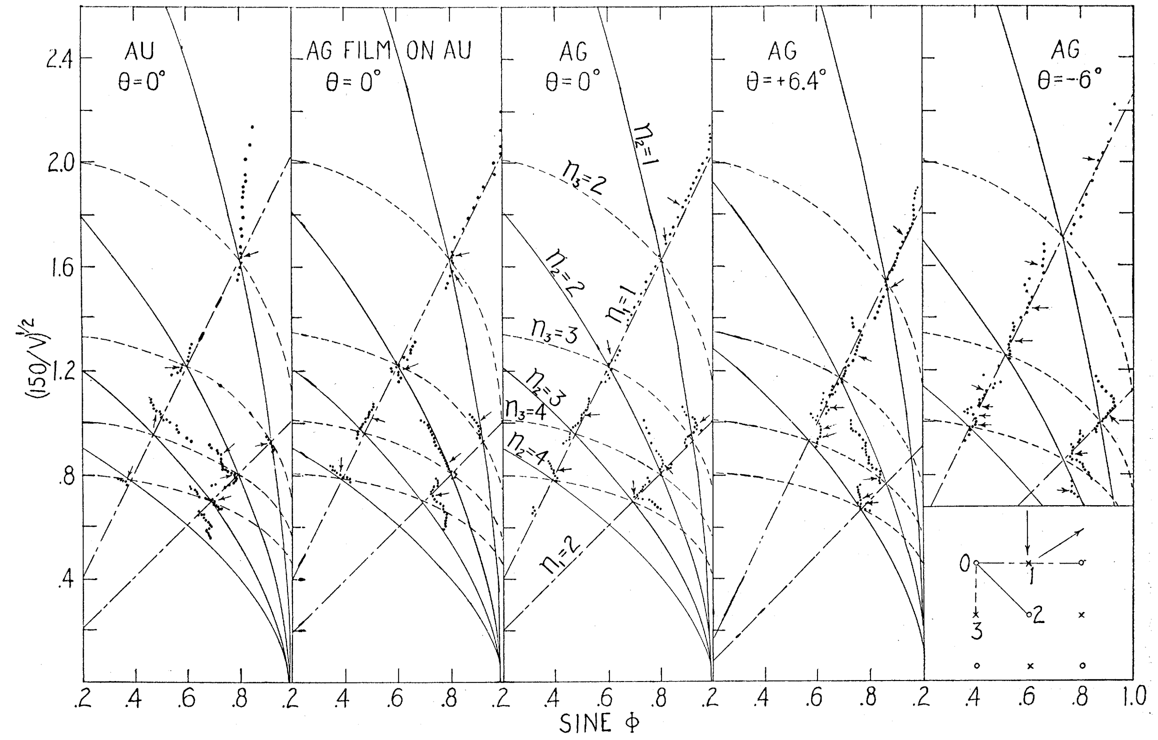

FIG. 4. Plots of $\sin \phi$ against $\lambda = (150/V)^{\frac{1}{2}}$. ϕ = colatitude angle measured from the normal. θ = angle of incidence of primary beam. The points are experimental observations, and the arrows indicate which ones correspond to maxima. The lines are obtained from grating formulas obtained by considering the corresponding rows of atoms as indicated in the lower right corner.

formulas are indicated in the lower right corner of the figure. Experimental points are given not only for voltages corresponding to maximum intensities, but at small voltage intervals over the range for which the beams can be detected during their growth and decay. It is seen that in general the surface grating formula is more nearly followed for the low-voltage beams, while a depth grating formula is a better approximation for the higher-voltage beams. But, in addition, it is to be noted that while the 3rd beam, 1st order, (3–1), follows the depth grating for the gold crystal, this beam follows the surface grating for both the silver crystal and the silver film on the gold crystal. Hence, if we grant that the grating irregularities and consequently the number of lines in the diffracting lattices are the same for the gold crystal and the silver film on the gold crystal, it appears necessary to conclude that some other factor is responsible for the difference in the behavior of this beam in the case of gold and silver crystals. One possibility is that the deviations from the surface grating formula result from the same mechanism as the fine structure of the diffraction beams. If this is the case there should be some correlation between fine structure and deviation from the surface grating formula. Both have been found to change with changes in the angle of incidence, but changes in fine structure characteristics are not always accompanied by changes in the deviation from the surface grating formula.

The two sets of curves at the right in Fig. 4 show the results for two different angles of incidence in the case of the (100) face of a silver crystal. It is to be noted that the experimental points for some of the beams follow one grating formula for one angle of incidence and a different grating formula for another angle of incidence. This is, however, not true for all of the beams. It is also seen that some of the experimental points for a beam may follow one grating formula while the remainder of the points for the same beam may follow another grating formula. These results suggest that under certain conditions some one direction in the crystal is more effective in producing constructive interference than other directions, a condition which might be realized by an unsymmetrical distribution of the potential in the outer regions of the atoms. One must, however, not overlook the possibility that if a portion of the surface is etched parallel to a set of atomic planes other than the one coinciding with the geometric boundary, this may be expected to cause the portions of the beam reflected from this surface to follow a grating formula for this surface.[4] But, it is not easy to see why all of the diffraction beams should not be equally affected. From the observations available this view does not appear sufficient to account for all of these characteristics. More observations are being made on this point.

The results of this investigation clearly show that the fine structure of the diffraction beams are determined at least to some extent by the nature of the atoms composing the crystal. It would seem that an extension of Morse's treatment[5] with fewer approximations is required to account for these results.

The purchase of the gold and some of the apparatus was made possible by a Grant-in-Aid from the National Research Council.

Messrs. K. P. Page and E. C. Bray have aided in taking the numerous observations.

Note added in proof: W. T. Sproull has now published a more detailed account of his investigation than that referred to above, and the writer is submitting some comments in a separate communication.

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[4] W. T. Sproull has recently reported results [Phys. Rev. **42**, 904 (1932)] from the (112) surface plane of a tungsten crystal. In the azimuth perpendicular to the cube diagonals beams which obey a depth grating formula corresponding to rows of atoms in the (011) plane were found at every primary voltage tried. In all probability this is because the etched surface facets of the crystal are parallel to the (011) plane rather than the (112) plane. Etching parallel to the more densely populated (011) plane would result from evaporation by prolonged heat treatment even though the etched surface was originally parallel to the (112) plane. This, of course, must be taken into account in evaluating the inner potential from such observations.

[5] P. M. Morse, Phys. Rev. **35**, 1310 (1930).