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DIFFRACTION OF ELECTRONS BY A CRYSTAL OF NICKEL

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ABSTRACT

The intensity of scattering of a homogeneous beam of electrons of adjustable speed incident upon a single crystal of nickel has been measured as a function of direction. The crystal is cut parallel to a set of its {111}-planes and bombardment is at normal incidence. The distribution in latitude and azimuth has been determined for such scattered electrons as have lost little or none of their incident energy.

Electron beams resulting from diffraction by a nickel crystal.—Electrons of the above class are scattered in all directions at all speeds of bombardment, but at and near critical speeds sets of three or of six sharply defined beams of electrons issue from the crystal in its principal azimuths. Thirty such sets of beams have been observed for bombarding potentials below 370 volts. Six of these sets are due to scattering by adsorbed gas; they are not found when the crystal is thoroughly degassed. Of the twenty-four sets due to scattering by the gas-free crystal, twenty are associated with twenty sets of Laue beams that would issue from the crystal within the range of observation if the incident beam were a beam of heterogeneous x-rays, three that occur near grazing are accounted for as diffraction beams due to scattering from a single {111}-layer of nickel atoms, and one set of low intensity has not been accounted for. *Missing beams* number eight. These are beams whose occurrence is required by the correlations mentioned above, but which have not been found. The intensities expected for these beams are all low.

The spacing factor concerned in electron diffraction by a nickel crystal.—The electron beams associated with Laue beams do not coincide with these beams in position, but occur as if the crystal were contracted normally to its surface. The spacing factor describing this contraction varies from 0.7 for electrons of lowest speed to 0.9 for electrons whose speed corresponds to a potential difference of 370 volts.

Equivalent wave-lengths of the electron beams may be calculated from the diffraction data in the usual way. These turn out to be in acceptable agreement with the values of h/mv of the undulatory mechanics.

Diffraction beams due to adsorbed gas are observed except when the crystal has been thoroughly cleaned by heating. Six sets of beams of this class have been found; three of these appear only when the crystal is heavily coated with gas; the other three only when the amount of adsorbed gas is slight. The structure of the gas film giving rise to the latter beams has been deduced.

THE investigation reported in this paper was begun as the result of an accident which occurred in this laboratory in April 1925. At that time we were continuing an investigation, first reported in 1921,¹ of the distribution-in-angle of electrons scattered by a target of ordinary (poly-

¹ Davisson & Kunsman, Science 64, 522, (1921).

crystalline) nickel. During the course of this work a liquid-air bottle exploded at a time when the target was at a high temperature; the experimental tube was broken, and the target heavily oxidized by the inrushing air. The oxide was eventually reduced and a layer of the target removed by vaporization, but only after prolonged heating at various high temperatures in hydrogen and in vacuum.

When the experiments were continued it was found that the distribution-in-angle of the scattered electrons had been completely changed. Specimen curves exhibiting this alteration are shown in Fig. 1. These curves are all for a bombarding potential of 75 volts. The electron beam is incident on the target from the right, and the intensities of scattering in different directions are proportional to the vectors from the point of bombardment to the curves. The upper curves (for different angles of incidence) are characteristic of the target prior to the accident. They are of the type

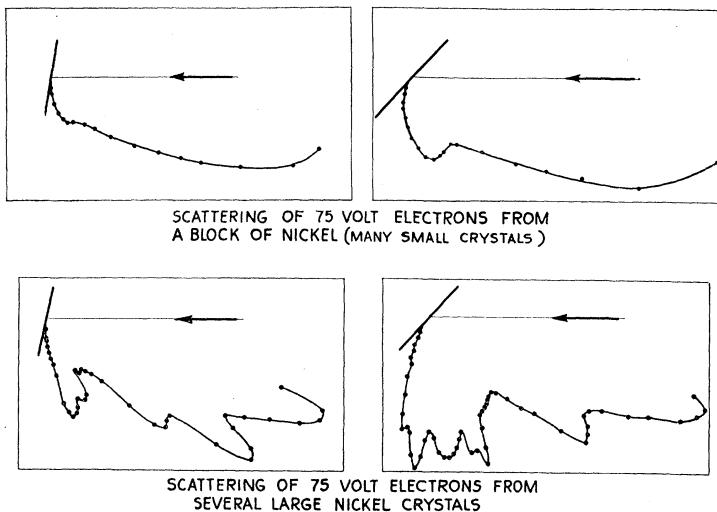


Fig. 1. Scattering curves from nickel before and after crystal growth had occurred.

described in the note in "Science" in 1921, and are similar to curves that have been obtained for nickel in four or five other experiments. The lower curves—obtained after the accident—were the first of their sort to be observed. This marked alteration in the scattering pattern was traced to a re-crystallization of the target that occurred during the prolonged heating. Before the accident and in previous experiments we had been bombarding many small crystals, but in the tests subsequent to the accident we were bombarding only a few large ones. The actual number was of the order of ten.

It seemed probable from these results that the intensity of scattering from a single crystal would exhibit a marked dependence on crystal direction, and we set about at once preparing experiments for an investigation of this dependence. We must admit that the results obtained in these experiments have proved to be quite at variance with our expectations. It seemed to us likely that strong beams would be found issuing from the crystal along what

may be termed its transparent directions—the directions in which the atoms in the lattice are arranged along the smallest number of lines per unit area. Strong beams are indeed found issuing from the crystal, but only when the speed of bombardment lies near one or another of a series of critical values, and then in directions quite unrelated to crystal transparency.

The most striking characteristic of these beams is a one to one correspondence, presently to be described, which the strongest of them bear to the Laue beams that would be found issuing from the same crystal if the incident beam were a beam of x-rays. Certain others appear to be analogues, not of Laue beams, but of optical diffraction beams from plane reflection gratings—the lines of these gratings being lines or rows of atoms in the surface of the crystal. Because of these similarities between the scattering of electrons by the crystal and the scattering of waves by three- and two-dimensional gratings a description of the occurrence and behavior of the electron diffraction beams in terms of the scattering of an equivalent wave radiation by the atoms of the crystal, and its subsequent interference, is not only possible, but most simple and natural. This involves the association of a wave-length with the incident electron beam, and this wave-length turns out to be in acceptable agreement with the value h/mv of the undulatory mechanics, Planck's action constant divided by the momentum of the electron.

That evidence for the wave nature of particle mechanics would be found in the reaction between a beam of electrons and a single crystal was predicted by Elsasser² two years ago—shortly after the appearance of L. de Broglie's original papers on wave mechanics. Elsasser believed, in fact, that evidence of this sort was already at hand in curves, published from these Laboratories,³ showing the distribution-in-angle of electrons scattered by a target of polycrystalline platinum. We should like to agree with Elsasser in his interpretation of these curves, but are unable to do so. The maxima in the scattering curves for platinum are of the type of the single maximum in the curves for nickel shown in the upper half of Fig. 1, and are, we believe, unrelated to crystal structure.

Preliminary announcement of the main results contained in this paper was made in "Nature" for April 16, 1927. In the present article we give a more complete account of the experiments and additional data.

THE APPARATUS

The essential parts of the special apparatus, Fig. 2, used in these experiments are the "electron gun" G , the target T and the double Faraday box collector C . The electrons constituting the primary beam are emitted thermally from the tungsten ribbon F , and are projected from the gun into a field-free enclosure containing the target and collector; the outer walls of the gun, the target, the outer box of the collector and the box enclosing these parts are held always at the same potential. The beam of electrons

² W. Elsasser, *Naturwiss.* **13**, 711 (1925).

³ Davisson & Kunsman, *Phys. Rev.* **22**, 242 (1923).

meets the target at normal incidence. High speed electrons scattered within the small solid angle defined by the collector opening enter the inner box of the collector, and from thence pass through a sensitive galvanometer. Electrons leaving the target with speeds appreciably less than the speed of the incident electrons are excluded from the collector by a retarding

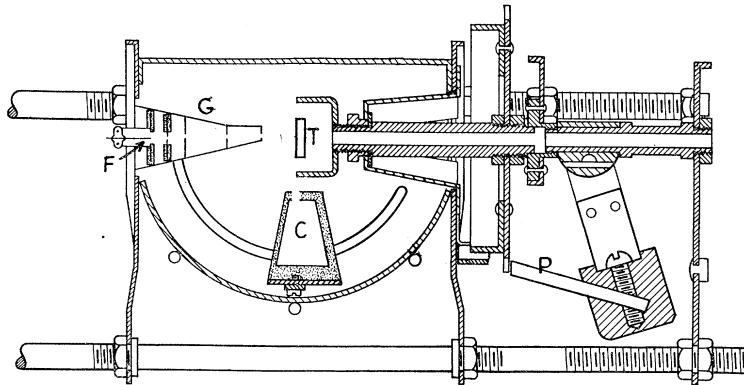


Fig. 2. Cross-sectional view of the experimental apparatus—glass bulb not shown.

potential between the inner and outer boxes. The angle between the axis of the incident beam and the line joining the bombarded area with the opening in the collector can be varied from 20 to 90 degrees. Also the target can be rotated about an axis that coincides with the axis of the incident beam. It is thus possible to measure the intensity of scattering in any direction in

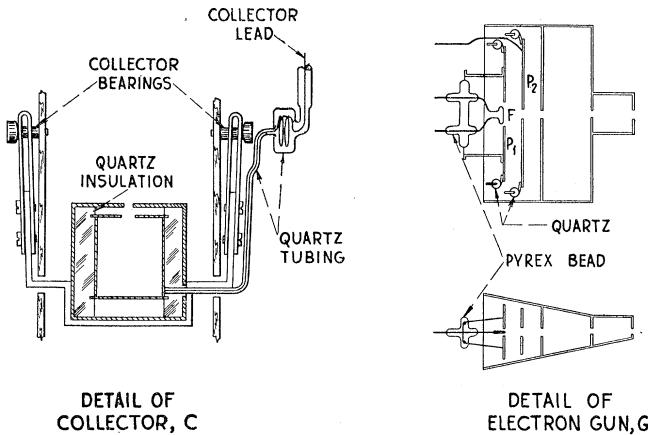


Fig. 3.

front of the target with the exception of directions lying within 20 degrees of the incident beam.

Details of the "electron gun" are shown in Fig. 3. The tungsten ribbon *F* lies in a rectangular opening in a nickel plate *P*₁. The purpose of this plate is to assist in concentrating the emission from the filament onto an opening

in the parallel plate P_2 . This is accomplished by making the potential of P_1 slightly more negative than that of the filament. The potential of P_2 relative to that of the filament is adjusted ordinarily to a rather high positive value.

The opening in P_2 is circular and slightly more than 1 mm in diameter. Some of the electrons passing through this opening continue on through apertures in a series of three plates that are at the same potential as the outer walls of the gun. It is the difference between this potential and that of the filament which determines the speed of the emergent beam. The first two of these apertures are 8 mm apart and are 1 mm in diameter; the diameter of the third is slightly greater. The geometry of these parts is such as to insure a well defined emergent beam relatively free from low speed secondary electrons. The gun was tested in a preliminary experiment, and was found to give a homogeneous beam. The distance from the end of the gun to the target is 7 mm.

The two parts of the collector (Fig. 3) are insulated from one another by blocks of clear quartz. The openings in the outer and inner boxes are circular,

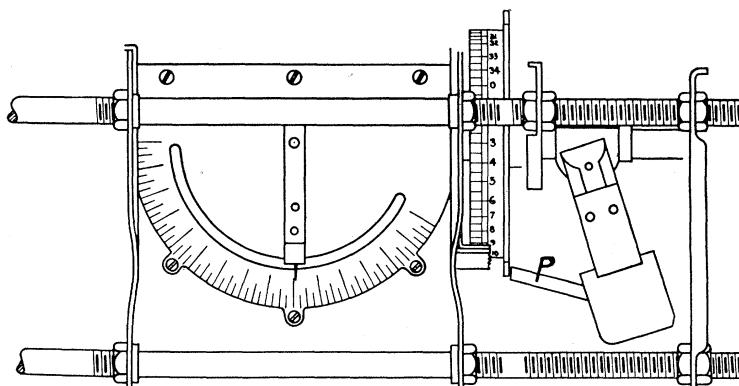


Fig. 4. Outside view of the experimental apparatus—glass bulb not shown—0.7 actual size.

their diameters being 1.0 mm and 2.0 mm respectively. These openings were made as near as possible to the side of the box adjacent to the gun in order to reduce to a minimum the unexplored region about the incident beam. The collector is suspended by arms from bearings outside the enclosing box (Fig. 4), and is free to rotate about a horizontal axis through the bombarded area and normal to the incident beam. The angular position of the collector is varied by rotating the whole tube, which is sealed from the pumps, about this axis. The lead to the inner box must be especially shielded from stray currents; it is enclosed in small quartz tubing from the point at which it leaves the outer box to the seal at which it leaves the tube. The distance from the bombarded area to the opening in the outer box is 11 mm.

The target is a block of nickel $8 \times 5 \times 3$ mm cut from a bar in which crystal growth had been induced by straining and annealing. The orientations of the largest crystals in the bar were determined by an examination of the optical reflections from crystal facets that had been developed by

etching. A cut was then made through one of the crystals approximately parallel to a set of its $\{111\}$ -planes. One of the surfaces so exposed was polished, etched, examined and corrected, and became eventually the face of the target.

No particular care was taken in preparing the target to avoid straining or damaging the crystal. The cutting was done with a jeweler's saw; holes were drilled through the ends of the block, and nickel wires were passed through these to serve as supports. After this rather rough usage the target was heated in an auxiliary tube to near its melting point without its showing any indication of recrystallization.

The effect of etching a nickel crystal, either chemically or by vaporization, is to develop its surface into sets of facets parallel to its principal planes. Those parallel to the $\{111\}$ -planes are developed most readily, but we have also observed others parallel to the $\{110\}$ -planes.⁴ Four sets of the predominant $\{111\}$ -facets are, in general, exposed on a plane surface. If one of these is parallel to the general plane of the surface, as in the case of our

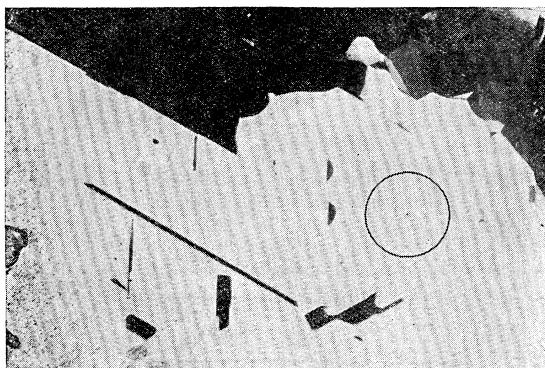


Fig. 5. Microphotograph of the nickel target.

target, the other sets have normals lying 20 degrees above the general plane of the surface and equally spaced in azimuth about the normal to the first set.

A microphotograph of the face of the target is shown in Fig. 5. The illumination is at normal incidence, and the large crystal shows white on account of the strong reflection from the $\{111\}$ -facets lying in its surface. That these facets make up nearly the whole of the surface seems probable from the fact that in the visual examination of the crystal the reflections from the lateral facets were very weak. This conclusion may not, however, be stated without reservation, as the weakness of these reflections may indicate merely that some dimension of the individual facets is small compared with optical wave-lengths. The regions appearing black in the photograph are made up of crystals having no facets parallel to the surface. Those included in the large crystal and others adjacent to it are twinned with the main structure. The area selected for bombardment is shown enclosed in a circle.

⁴ See also Potter and Sucksmith, Nature 119, 924 (1927), who found $\{100\}$ -facets.

The target was mounted in a holder from which it was insulated, and the holder was fixed to the end of a hollow shaft mounted in bearings. A small tungsten filament mounted back of the target (not shown in Fig. 2) supplies electrons for heating the target by bombardment. Leads from the target and from this filament pass out through the hollow shaft and are connected through platinum brush contacts to other leads which are carried through seals in the tube.

The mechanism for rotating the target is shown in Figs. 2 and 4. When the tube is rotated counter-clockwise about the collector axis, to bring the collector into range in front of the target, the molybdenum plunger *p* (attached to a heavy pendulum) passes through an opening in a toothed wheel (attached to the shaft) and engages with a milled edge of a strip of molybdenum that is attached to the frame. The wheel and the target are then locked to the frame. When the tube is rotated clockwise until the main or longitudinal axis of the tube has passed slightly beyond the horizontal, the plunger disengages from the milled edge but still remains within the opening in the toothed wheel. The pendulum has a second degree of freedom (it revolves about a fixed hollow shaft coaxial with the shaft carrying the target) so that, by rotating the tube about its main axis, the pendulum and engaged wheel are rotated relative to the frame. The range of this rotation is only 20° or 30°, but by rotating the tube slightly further in the clockwise direction about the collector axis the plunger is disengaged from the wheel, and can be moved, by rotation again about the main axis, to a different opening in the toothed wheel. By these operations the target can be worked through any angle. Its azimuth is read from a scale ruled on the wheel. This scale and that for reading the position of the collector are shown in Fig. 4. The bearings throughout the tube, with the exception of one nickel on nickel bearing, are either molybdenum on molybdenum, or molybdenum on nickel.

PREPARATION OF THE TUBE

The metal parts of the tube were preheated to 1000°C in a vacuum oven, and were then assembled and sealed into the bulb with the least possible delay. The bulb is of Pyrex and has sealed to it two auxiliary tubes, one containing cocoanut charcoal, and the other a misch metal vaporizer. This latter consists of a small pellet of misch metal attached to a molybdenum plate anode which may be bombarded from a nearby tungsten filament. The thermal contact between the pellet and the plate is reduced by the interposition of a narrow strip of molybdenum, so that the misch metal may be vaporized only by raising the plate to a very high temperature. The misch metal is vaporized when the pumping is nearly completed, and various of its constituents form solid compounds with the residual gas, thus improving and maintaining the vacuum.

During the pumping, which lasted several days, the tube itself and the tubing connecting it with the pumps were baked for hours at a time at 500°C, and the side tube containing charcoal was baked at an even higher temperature—about 550°C. This baking was alternated with heating by bom-

bardment of such of the metal parts as could be reached from the filaments. The target in particular was heated several times to a temperature at which it vaporized freely. The tube was sealed from the pumps with the target at a high temperature, and the charcoal at 400 or 500°C and cooling. The pressure in the tube at the time was 2 or 3×10^{-6} mm of mercury. As soon as the tube containing charcoal had cooled sufficiently it was immersed in liquid air. No means were provided for measuring the pressure of the gas in the tube after sealing from the pumps, but from experience with similar tubes in which such measurements could be made we judge that its equilibrium value was 10^{-8} mm of mercury or less. The pumping equipment consisted of a three stage Gaede diffusion pump backed by a two stage oil pump.

THE CRYSTAL

It is important to have a clear picture of the arrangement of atoms presented to the incident beam by the crystal. The nickel crystal is of the

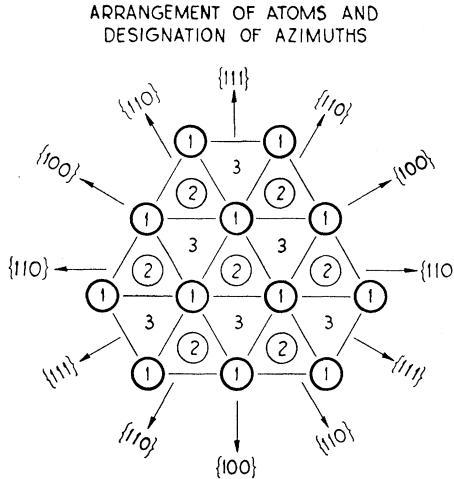


Fig. 6.

face-centered cubic type. The $\{111\}$ -plane is the plane of densest packing, and in this plane the atoms have a triangular arrangement. Looking directly downward onto a crystal cut to this plane (Fig. 6) one sees the atoms of the second plane below the centers of alternate triangles of the first plane, and the atoms of the third plane below the centers of the remaining triangles. The atoms of the fourth plane are below those of the first. The lines joining any second-layer atom with the three nearest first-layer atoms are $\{110\}$ -directions in the crystal, and the lines joining it with the three next-nearest surface atoms are the orthogonal $\{100\}$ -directions. It will be convenient to refer to the azimuths of these latter directions as $\{100\}$ -azimuths. The azimuths of the $\{110\}$ -directions are also those of the three lateral $\{111\}$ -directions, already referred to, and we shall designate these as $\{111\}$ -azimuths. We need also a designation for the azimuths that bisect the dihedral angles between adjacent members of the two sets already specified. There are six such azimuths and they will be referred to as $\{110\}$ -azimuths.

It follows from the trigonal symmetry of the crystal that if the intensity of scattering exhibits a dependence on azimuth as we pass from a $\{100\}$ -azimuth to a next adjacent $\{111\}$ -azimuth (60°), the same dependence must be exhibited in the reverse order as we continue on through 60° to the next following $\{100\}$ -azimuth. Dependence upon azimuth must be an even function of period $2\pi/3$.

DISTRIBUTION OF SPEEDS AMONG SCATTERED ELECTRONS

The electrons leaving the target in any given direction appear always to have speeds that are distributed in one of two ways, depending upon whether the direction lies within or outside a diffraction beam. In the latter

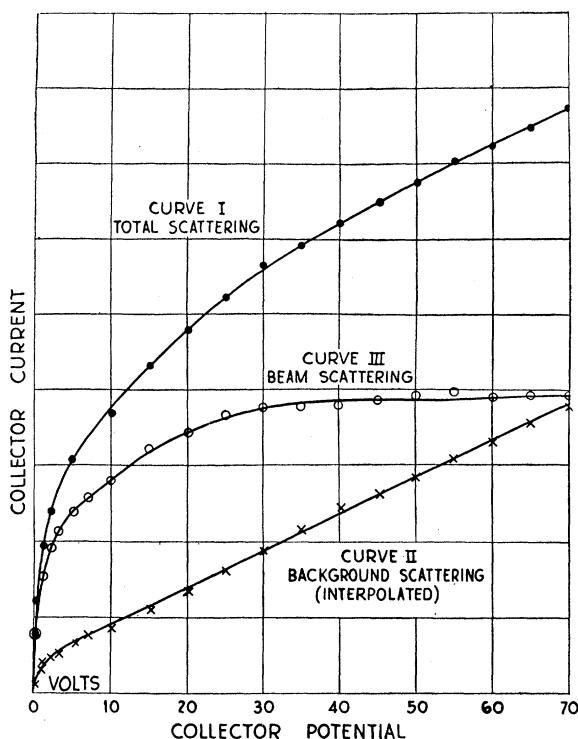


Fig. 7. Current to collector as a function of collector potential—bombarding potential 160 volts.

case—that of the electrons making up the “background” scattering—there is always a definite group having the speed of those in the incident beam. Below this speed there is in general a range over which the distribution in energy is very nearly uniform; and below this a range, ending with zero speed, in which the representation increases rapidly with decreasing energy.

These characteristics are inferred from the relation between collector current and the potential of the collector relative to that of the filament. The lower portion of a typical curve exhibiting this relation for the “background” scattering is shown in Fig. 7 (Curve II). The ordinate of a curve

of this sort is not, of course, an actual measure of the number of electrons entering the outer box with sufficient energy to reach the inner box. On account of the distortion of the field about the openings the probability that an electron entering the outer box with just sufficient energy to reach the walls of the inner box will actually do so is vanishingly small; the saturation current due to electrons of a given speed is attained only when the potential of the inner box is somewhat higher than that corresponding to their speed. The rounding off of the current-voltage curve at the top of the initial rise is due to some extent to this cause.

For this reason the group of high speed, or full speed, electrons is more nearly homogeneous than would be inferred from the current-voltage curve if no account were taken of this distortion. It seems probable, in fact, that the distortion accounts almost completely for the rounding off of the curve, and that the group is as nearly homogeneous as is permitted by the drop in potential along the filament and the initial speeds of the emitted electrons. This view is strongly supported by the observations of Becker,⁵ Brown and Whiddington,⁶ Sharman,⁷ and Brinsmade⁸ on the magnetic spectrum of electrons scattered by metals, and by similar curves obtained by Farnsworth.⁹

Within a diffraction-beam the distribution-in-speed is somewhat different. There is again a definite group of full speed electrons, but speeds just inferior to the maximum have much greater representation than among the background electrons. This is inferred from curve III in Fig. 7 which is representative of the current-voltage relation for electrons of this class. We shall return in a later section to a further consideration of the curves in this figure.

In studying the distribution in direction of the scattered electrons measurements have been confined, as nearly as possible, to the group of full speed electrons. The potential of the collector is set just high enough to admit all of this group. The ratio of collector to bombarding current is then of the order 10^{-4} , so that, by using bombarding currents of the order 10^{-6} ampere, collector currents are obtained that are easily measurable with a sensitive galvanometer. The total integrated current of full speed scattered electrons is from a tenth to a twentieth as great as the current of the incident beam.

DISTRIBUTION OF DIRECTIONS AMONG FULL SPEED SCATTERED ELECTRONS

The current of full speed electrons entering the collector is proportional to the current incident upon the target and is otherwise a function of the bombarding potential and of the latitude and azimuth of the collector. Three simple types of measurement are thus possible in each of which two of the independent variables are held constant and the third is varied. When bombarding potential and azimuth are fixed and exploration is made in

⁵ J. A. Becker, Phys. Rev. **23**, 664 (1924).

⁶ D. Brown and R. Whiddington, Nature **119**, 427 (1927).

⁷ C. F. Sharman, Proc. Camb. Phil. Soc. **23**, 523 (1927).

⁸ J. B. Brinsmade, Phys. Rev. **30**, 494 (1927).

⁹ H. E. Farnsworth, Phys. Rev. **25**, 41 (1925).

latitude a dependence of current upon angle is observed which is generally of the form shown in Fig. 8; the current of scattered electrons is zero in the plane of the target and increases regularly to a highest value at the limit of observations—colatitude 20° . This type of dependence upon angle is essentially the same as is observed when the target is of ordinary nickel—made up of many small crystals.

When bombarding potential and latitude angle are fixed and exploration is made in azimuth a variation of collector current is always observed, and this exhibits always the three-fold symmetry required by the symmetry of the crystal. The curves show in general two sets of maxima—a set of three in the $\{111\}$ -azimuths, and a set of three of different intensity in the $\{100\}$ -azimuths. These crests and troughs in the azimuth curves are usually not pronounced.

In the third method of observation the position of the collector is fixed in one of the principal azimuths at one after another of a series of colatitude

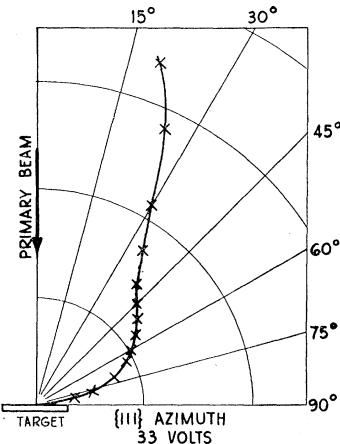


Fig. 8. Typical colatitude scattering curve for the single nickel crystal.

angles, and at each such setting the current to the collector is observed as a function of bombarding potential. It would be desirable in making observations of this sort to keep constant the current in the incident beam but, as there is no ready means of doing this, the current to the plate P_2 (Fig. 3) is kept constant instead. Beginning at colatitude 20° a series of such observations is made, over a predetermined voltage range, at 5° intervals to colatitude 80° or 85° . A portion of a set of curves constructed from such data is shown in Fig. 9.

The general trend of a single one of these curves is not significant as it is determined in part by variation with voltage of the bombarding current. The relative displacements among them, however, are significant as they indicate departures from the simple type of colatitude curve shown in Fig. 8. From the curves in Fig. 9 we see, for example, that the colatitude curves for bombarding potentials near 55 volts are characterized by exceptional intensities at colatitude angles near 50° . The data for constructing colatitude

curves for particular bombarding potentials are taken directly from such curves as those of Fig. 9, or the features in these latter curves are used as

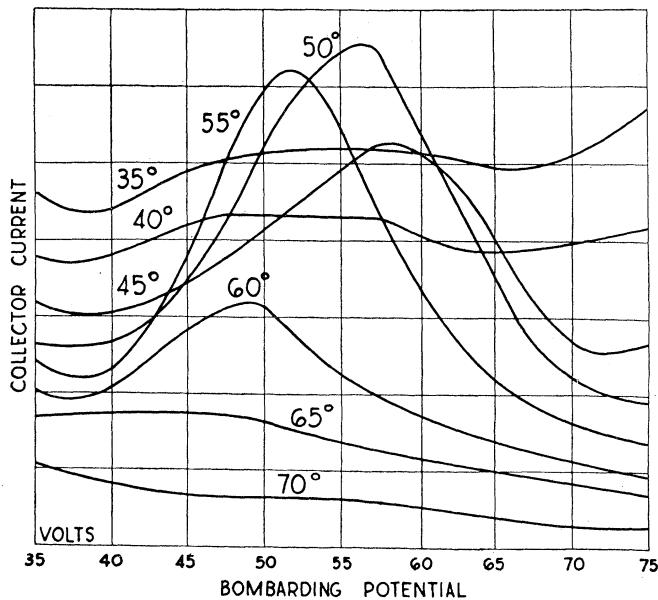


Fig. 9. Curves of collector current vs. bombardment potential—showing the development of the "54 volt beam." Azimuth {111}.

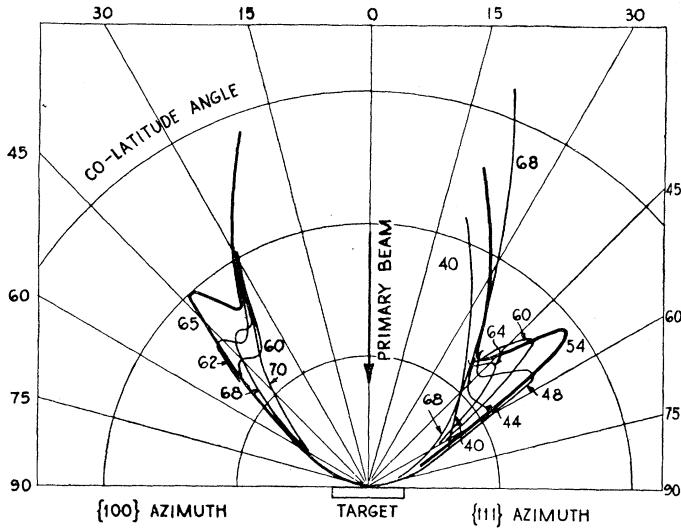


Fig. 10. Scattering curves showing the occurrence of the "54 volt" electron beam and the "65 volt" electron beam. (On each scattering curve is indicated the bombarding potential in volts.)

a guide to voltage-colatitude ranges requiring special study. This method has been employed in exploring the principal azimuths in the range from

15 to 350 volts, and two other azimuths, to be specified later, over the range from 200 to 300 volts. Every feature of the sort shown in Fig. 9 has been investigated.

The unusual and significant feature revealed by the curves of Fig. 9 is exhibited again in the set of colatitude curves on the right in Fig. 10. We see a slight hump at 60° in the colatitude curve for 40 volts, and observe that as the bombarding potential is increased this hump develops into a strong spur which reaches a maximum development at 54 volts in colatitude

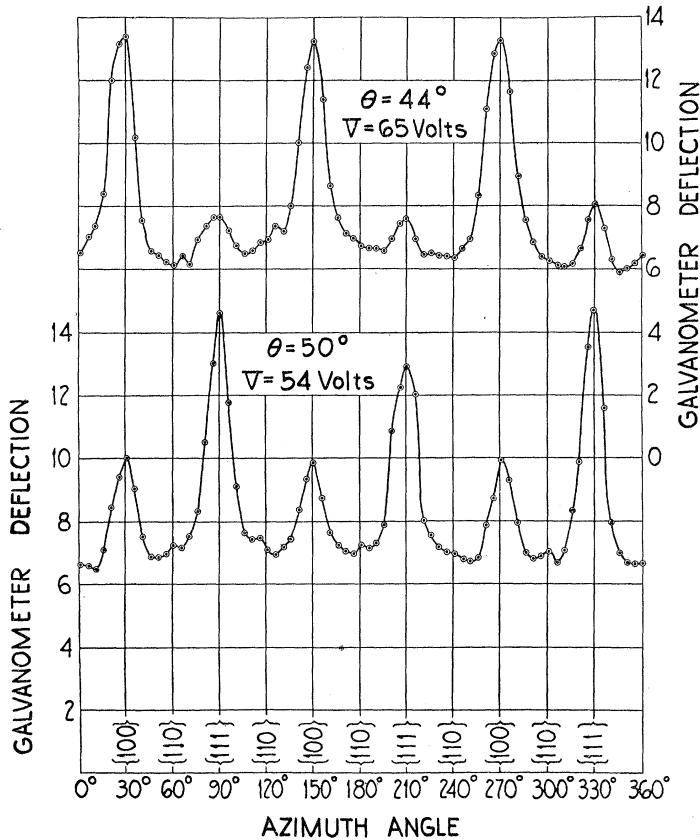


Fig. 11. Azimuth scattering curves through the "54-volt" electron beam and through the "65-volt" electron beam.

50° , then decreases in intensity and finally disappears at about 66 volts in colatitude 40° .

A section in azimuth through the center of this spur in its maximum development is shown in the lower curve of Fig. 11. The spur is sharp in azimuth as well as in latitude and is one of a set of three spurs as the symmetry of the crystal requires. The smaller peaks in the $\{100\}$ -azimuth are sections of a similar set of spurs that attains its maximum development at 65 volts in colatitude 44° . A complete set of colatitude curves

for one of these spurs is shown on the left in Fig. 10. The upper curve in Fig. 11 is an azimuth curve through the peaks of these 65 volt spurs. The small peaks in the {111}-azimuth are the remnants of the "54 volt" spurs.

The colatitude angles at which the various spurs of a single set are strongest are found not to have exactly the same values. This is due apparently to imperfect alignment of the normal to the crystal planes and the axis of rotation of the target. In each of several sets that have been studied these angles are expressed by the formula $\theta = \theta_0 + \Delta\theta \cos(\phi - \phi_0)$, where θ_0 is a constant for a given set and is taken to represent the colatitude angle at which all spurs in the set would be strongest if the alignment were perfect, and $\Delta\theta$ and ϕ_0 are constants that have the same values for all sets, 2° and 1° respectively. This is taken to mean that the axis of rotation is displaced about one degree from the normal to the crystal planes into azimuth 181° . The correction $2 \cos(\phi - 1^\circ)$ degrees has been applied to all observed values of the colatitude angle θ .

The voltages at which the different spurs of a given set are strongest probably show a like variation. The differences are slight, however, and no attempt has been made to apply a voltage correction.

If we regard the spur as a feature superposed on the simple scattering curve the position of its maximum is falsified to some extent by the variation with angle of the background against which it appears. The method of correcting for this effect is indicated by the curves in Fig. 12. The end portions of the observed curve are joined by a curve of the known form of the simple relation (see e.g., Fig. 8), and the difference of these curves is plotted as the graph of the spur. The position of the maximum of this difference curve is taken as the true value of θ .

THE WIDTH OF THE SPUR

From the difference curve in Fig. 12 we see that the spur has an apparent angular width of about 25° . What width is to be expected if the spur is due to a beam of electrons which is as sharply defined as the primary beam? This latter beam is defined by circular apertures 1 mm in diameter, and if we assume that the beam is a cylinder of this diameter, an equally sharp beam scattered at colatitude θ would extend over a colatitude arc of $(1 \times \cos \theta)$ mm. The circular opening in the outer box is 1 mm in diameter, and its distance from the axis of rotation is 11 mm, so that the least possible value for the apparent colatitude width is $(1 + \cos \theta)/11$ radians, or $5.2(1 + \cos \theta)$ degrees. For the spur under consideration $\theta = 50^\circ$, and the calculated width is 8.5° .

The width in azimuth of the same spur is seen from the lower curve of Fig. 11 to be about 30° . The least value for this width is given by $5.2(2)/\sin \theta$ which for $\theta = 50^\circ$ amounts to 13.5° . Thus both in colatitude and in azimuth the observed width of the beam is more than double the least possible value.

It is to be expected, of course, that the observed values will be somewhat greater than those calculated, since it is hardly likely that the primary beam is as sharply defined as has been assumed; it is probably divergent as well as somewhat nonhomogeneous. There is no way, however, of investigating

these matters. The most that can be said with certainty is that the spur is due to a beam of electrons whose definition is comparable in sharpness with that of the incident beam.

DISTRIBUTION OF SPEEDS AMONG ELECTRONS CONSTITUTING THE DIFFRACTION BEAMS

Assuming that the sharply defined beam is a distinct feature superposed upon the general background scattering, it is natural to inquire in what way

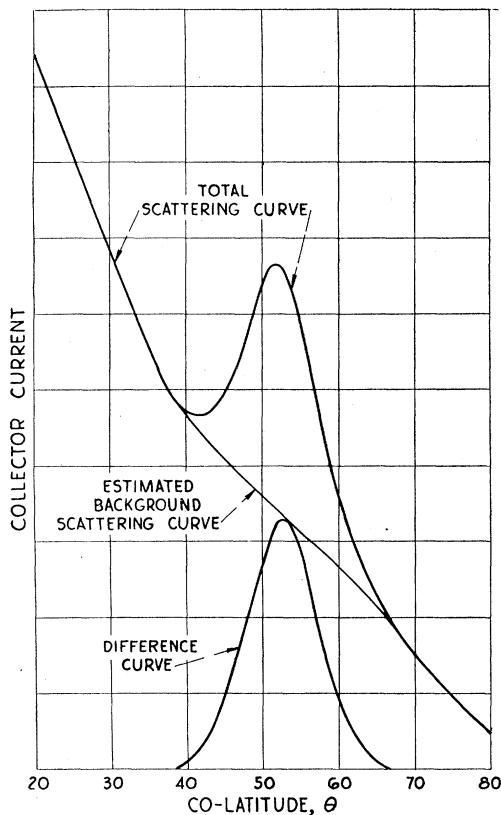


Fig. 12. Scattering curve of the "54 volt" beam, showing the method of determining the position of the maximum. (Angles not corrected for the tilt of the target).

the electrons constituting the whole of the superposed scattering are distributed in speed. Is the complete superposed scattering made up of full speed electrons only, or is it made up in part of electrons of lower speeds? This point has been investigated in several different ways. The fact seems to be that in addition to the group of full speed electrons observed in the distribution-in-direction measurements the complete superposed scattering includes also other electrons that have lost energy in various amounts up to about one quarter of their incident energy; it seems to include no electrons that have lost more than this amount.

These characteristics are inferred from the experimental results exhibited in Fig. 7. The upper curve was obtained with the collector in the center of a beam in the {100}-azimuth which is strongest at 160 volts. Two other curves (not shown) were then obtained with the position of the collector unaltered, but with the bombarding potential changed to 120 volts in one case, and to 200 volts in the other. The spur does not appear at either of these voltages, so that the current-voltage curves for these voltages are for "background" electrons only. These curves are not very different, so that a similar curve for 160 volts may be interpolated with considerable certainty. Curve II of Fig. 7 is this interpolated curve representing the current-voltage relation for "background" electrons only at 160 volts.

Curve III has been obtained by subtracting II from I, and consequently represents the current-voltage curve that would be obtained if the "background" scattering could be eliminated. We infer from the form of this curve that the electrons making up the whole of the superposed scattering are distributed in energy as described in the previous paragraph. The distribution is that which might be expected among the emergent electrons if a homogeneous beam were incident upon an extremely thin plate—a plate only one or two atoms in thickness.

Although the matters here considered require much more study than they have so far received, it is fairly clear that the superposed scattering is made up of beams of full speed and nearly full speed electrons that approximate the incident beam in sharpness, and that these beams appear only at and near certain critical bombarding potentials.

POSITIONS AND VOLTAGES OF ELECTRON BEAMS

The work of investigating these beams and searching out new ones has progressed in several distinct stages. To begin with an exploration was made through the principal azimuths in the range 15 to 200 volts. Thirteen sets of beams were found, and these were described in our note to "Nature." The exploration was then extended to 350 volts, and eight additional sets were found. Up to this time the target had been heated last while the tube was still on the pumps.

After completing the exploration to 350 volts the target was strongly heated, and allowed to cool again to room temperature. The effect of this treatment was to increase generally the intensities of the beams without altering either the voltages at which they occurred or their positions. Three sets of beams only were exceptions to this rule, and these were the particular three sets which in our note in "Nature" we regarded as anomalous; the intensities of these were decreased.

These alterations in intensity resulted, we believe, from removal of gas from the surface of the target. In further tests it was found that immediately after bombardment, while the target is still hot, the beams are all weak; that they then increase in intensity as the target cools and that later, presumably as gas collects again on the surface, their intensities decrease. This final decrease in intensity was rather rapid after the first heat treatments, but after ten or a dozen heatings it was much slower—the intensities of the

beams remaining for hours near their maximum values. This was the behavior of the normal beams. The three sets of anomalous beams were progressively weakened, and finally disappeared.

The alteration brought about in the "54 volt" beam by this degassing of the target is shown in Fig. 13. Curve A is reproduced from Fig. 10 and shows the beam in its maximum development in the earlier tests. Curve B is for the same beam from the cleaned target. The intensity has been increased between four and five fold, while the intensity of the background scattering has been decreased. For beams of higher voltage the increase in intensity was in general less marked, but even for beams above 300 volts intensities were at least doubled. *The ratio of full speed electrons scattered into any one of the most intense sets of beams to the total number scattered in all directions is about two-tenths.*

A further effect of cleaning the target has been to cause the appearance of certain new beams in the range below 200 volts. These beams are of peculiar interest. They are exceptionally sensitive to gas, and were entirely absent at the time the earlier observations were made.

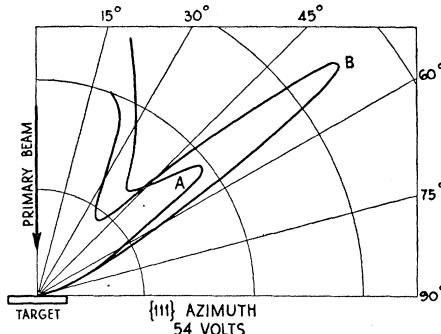


Fig. 13. The "54 volt" beam before and after heating the crystal by electron bombardment.
A. Original condition as in Fig. 10. B. After heating the crystal.

We have found altogether thirty sets of beams, including those due to gas. Most of these are analogues of Laue beams, and the data for beams of this class are listed in Table I. The intensities in column 5 are estimated for a constant electron current bombarding the target, and for the target surface as free as possible from gas. In a few cases redetermined constants for the beams of Table I are somewhat different from those given in the note in "Nature." The beams not listed in Table I will be considered individually.

It would be possible to follow, with the beams listed in Table I, the procedure employed in our note in "Nature" (to point out a correspondence between the electron beams and the Laue beams that would issue from the same crystal if the incident beam were a beam of x-rays); then, with this suggestion of the wave nature of the phenomenon, to show that wave-lengths may be associated in a simple and natural way with the electron beams, and finally to compute these wave-lengths and show that they are in accord with the requirements of the undulatory mechanics. It is prefer-

able, however, to start at once with the idea that a stream of electrons of speed v is in some way equivalent to a beam of radiation of wave-length h/mv , and to show to what extent the observations can be accounted for on this hypothesis. We assume that this radiation is scattered and absorbed by the atoms of the crystal and that, just as in the case of x-rays, strong diffraction beams result from coincidence in phase of all of the radiation scattered in some particular direction.

TABLE I
Space lattice electron beams

Azi.	Bombarding Potential V	Equivalent Wave-length $\lambda = (150/V)^{1/2}$	Colatitude θ	Beam Intensity (Arbitrary Scale)	Beam Int. Background Int.
{111}	54 volts	1.67 Å	50°	1.0	7.0
	106	1.19	28	0.4	1.4
	174	0.928	22	2.0	1.3
	181	0.910	55	0.7	1.0
	248	0.778	44	1.0	2.6
	258	0.762	<20	4.5	1.8
	343	0.661	34	3.0	1.5
{100}	347	0.657	62	0.07	0.3
	65	1.52	44	1.0	7.0
	126	1.09	28	2.0	3.8
	160	0.968	60	0.8	5.7
	190	0.889	20	2.0	1.3
	230	0.807	46	0.4	1.2
	292	0.716	<20	7.0	2.0
	310	0.695	70	0.15	0.8
{110}	312	0.693	37	1.5	1.2
	370	0.636	57	0.15	0.4
	143	1.024	56	0.2	0.9
	170	0.940	46	0.1	0.5
	188	0.893	43	0.3	1.0
	248	0.778	34	0.45	0.6

In considering the conditions under which such beams will occur it will be convenient to regard the crystal as built up of {111}-planes of atoms parallel to the principal facets, and to picture the radiation scattered by the crystal as made up of the contributions from all such planes. This viewpoint has a distinct advantage, in the present case, over regarding diffraction beams as built up of contributions regularly reflected from the Bragg atom-planes. The amplitude of the radiation proceeding in a given direction from the crystal is then to be regarded as the sum of the amplitudes (with due regard to phase) of the increments of radiation proceeding in the same direction from all such {111}-planes of atoms—or more precisely, the fractions of such increments that actually escape from the crystal.

If we imagine a system of Cartesian coordinates with its origin at the center of a surface atom, its positive z -axis extending outward from the facet, and its positive x -axis lying in one of the {110}-azimuths of the crystal, then atom centers occur at the points

$$\begin{aligned}x &= (M+N+P)s/2 \\y &= 3^{1/2}(M-N+P/3)s/2 \\z &= -2^{1/2}Ps/3\end{aligned}$$

where $s (=a/2^{1/2})$ represents the least distance between atoms in a $\{111\}$ -plane (the side of the elementary triangle), M and N are any integers, and P is zero or any positive integer.

If a beam of plane waves is incident upon any one of these atom planes along a line whose direction cosines are l_1, m_1, n_1 , it may be shown that the radiation scattered by the individual atoms will be in phase along the directions

$$\begin{aligned}l_2 &= l_1 + (p+r)\lambda/s \\m_2 &= m_1 + (p-r)\lambda/3^{1/2}s \\n_2 &= \pm(1-l_2^2-m_2^2)^{1/2}\end{aligned}$$

where λ represents the wave-length of the radiation, and p and r are any integers. If the waves meet the layer at normal incidence, as in our experiments, then $l_1 = m_1 = 0, n_1 = -1$, and

$$\begin{aligned}l_2 &= (p+r)\lambda/s \\m_2 &= (p-r)\lambda/3^{1/2}s \\n_2 &= \pm(1-l_2^2-m_2^2)^{1/2}\end{aligned}$$

It may be shown that these are just the directions in which diffraction beams are to be expected if the plane of atoms is regarded as equivalent to a great number of line gratings. All of the atoms in the plane may be regarded as arranged on lines parallel to the line joining any two of them, and every such set of lines functions as a line grating. Diffraction beams due to each such grating occur in the plane normal to its lines, and satisfy the ordinary plane grating formula, $n\lambda = d \sin \theta$.

The grating constant d has its greatest value for the three plane gratings the lines of which are parallel to the sides of the elementary triangle. For these $d = d_1 = 3^{1/2}s/2$, and $n\lambda = (3^{1/2}s/2)\sin \theta$. The beams due to these gratings occur symmetrically in the $\{111\}$ and $\{100\}$ -azimuths.

The longest wave-length that can give rise to a diffraction beam is found by setting $n = 1$ and $\sin \theta = 1$ in the grating formula in which d has its greatest value, i.e., it is the wave-length $3^{1/2}s/2$. When waves of this length are incident normally on the plane of atoms first order diffraction beams should appear at grazing emergence in the $\{111\}$ and $\{100\}$ -azimuths—six beams in all. When the wave-length is decreased these beams should split into two sets of six beams each—one set moving upward toward the incident beam, and the other set moving downward. We are concerned with the upward moving set only. When the wave-length has been decreased to $3^{1/2}s/4$ second order beams should appear at grazing, and these should follow the course of the first order beams with still further decrease in the wave-length. In the meantime, however, six first order beams from the three gratings of second largest spacing ($d = d_2 = s/2$) should have appeared at grazing in the $\{110\}$ -azimuths. And as the wave-length is shortened beams should appear in still other azimuths.

These are the diffraction beams to be expected from a single layer of atoms. In general, when the incident beam is being scattered simultaneously from a large number of such layers the diffraction beams from the different layers must emerge from the crystal out of phase with one another, and the amplitude of the resultant beam must be much smaller than that which a single layer should produce.

There are two conditions, however, under which the amplitude of a given set of beams may be as great as, or greater than, that due to a single layer. One of these is the well known case of the Laue beams in which the scattered waves contributed to a particular beam from the consecutive atom layers are in phase and reenforce one another by constructive interference; the other is the case in which the reduction in intensity of the radiation on passing through a single layer of atoms—due to scattering and absorption—is so great that no appreciable radiation emerges from the interior of the crystal to interfere with that scattered by the first layer. The resultant scattering in this case will be approximately that from a single layer of atoms. This condition will be most closely approached near grazing, since in this region the paths in the crystal over which radiation from the second and lower layers must escape are greatly lengthened. The new electron beams discovered below 200 volts after the crystal was thoroughly degassed appear to be of this latter type. We shall begin our discussion of the data with an examination of these “plane grating” beams.

The value of the spacing s for nickel is 2.48\AA , so that $d_1 = 3^{1/2}s/2 = 2.15\text{\AA}$, and first order beams should appear at grazing emergence in the $\{111\}$ and $\{100\}$ -azimuths when $\lambda (=h/mv)$ has this value. Rewriting de Broglie's formula for λ in terms of the kinetic energy V of the electrons expressed in equivalent volts, we have:

$$\lambda (\text{in Angstrom units}) = (150/V)^{1/2}$$

From this we calculate that the electron wave-length will have its critical value when $V = 32.5$ volts.

The new beams in the $\{100\}$ and $\{111\}$ -azimuths are shown in Figs. 14 and 15 in which current to the collector is plotted against bombarding potential for various colatitude angles. In both azimuths beams appear at grazing ($\theta = 90^\circ$) at or very near the calculated voltage, and then move upward toward the incident beam as the voltage is increased. The low intensities of the beams near grazing are attributed to the general roughness of the surface and to absorption of the radiation scattered by the first layer by the atoms of the same layer. The final disappearance of these beams at about 30° above the surface is accounted for by interference between the radiation scattered by the first layer and that escaping from below. Over this range we expect wave-length and angular position of the beam to be related through the plane grating formula $\lambda = d_1 \sin \theta$, or voltage and position through the equivalent relation $V^{1/2} \sin \theta = (32.5)^{1/2} = 5.70$. That the observations are in accord with this requirement is shown by the values calculated from Figs. 14 and 15 and recorded in Table II.

TABLE II
Occurrence of "plane grating" electron beams.

θ	Azimuth - $\{100\}$ V	Azimuth - $\{111\}$ V	Azimuth - $\{110\}$ V	
	$V^{1/2} \sin \theta$	$V^{1/2} \sin \theta$	$V^{1/2} \sin \theta$	
85	32.0	5.64	97.5	9.83
80	33.0	5.66	100.0	9.85
75	35.0	5.72	103.5	9.83
70	36.0	5.64	108.0	9.77
65	38.5	5.63	112.5	9.62
60	42.5	5.65		

The difference in intensity between the beams shown in Figs. 14 and 15 is due apparently to a real dependence of intensity upon azimuth; the beams in the three $\{100\}$ -azimuths are all more intense than the beams in the $\{111\}$ -azimuths. We naturally try to account for this difference (which could not occur if the scattering were from a single atom layer) by supposing that although the extinction of the radiation in the metal is sufficiently great to leave first layer scattering predominant, it is not sufficiently great, even

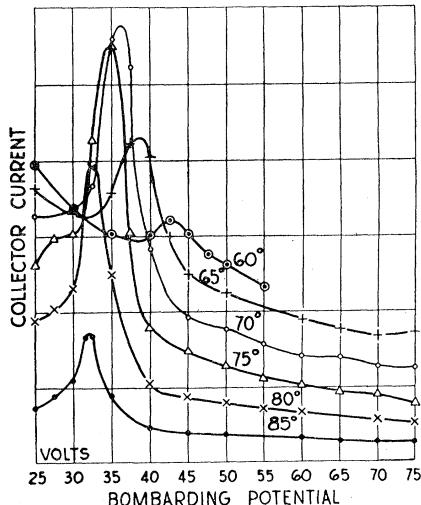


Fig. 14. Collector current vs. bombarding potential showing plane grating beams near grazing in $\{100\}$ -azimuth.

near grazing, to suppress completely the escape of radiation from lower layers. The phase difference between first and second layer beams is not the same in the two azimuths, and as a consequence an intensity difference results. Whether the observed difference is in the sense to be expected will be considered later.

It was expected that second order beams corresponding to the ones just described would be found at grazing for $V = 4 \times 32.5 = 130$ volts. These appear, however, to be entirely missing. We cannot account for this.

On the other hand the anticipated first order beams in the $\{110\}$ -azimuths resulting from the atomic line gratings of second widest spacing are duly found. One of these beams is shown in Fig. 16. The grating constant is $s/2$,

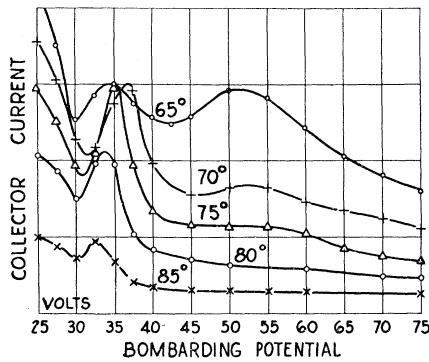


Fig. 15. Collector current vs. bombarding potential showing plane grating beams near grazing in $\{111\}$ -azimuth.¹⁰

or $1/3^{1/2}$ as great as that of the former gratings, so that the beam should appear at grazing for $V=3 \times 32.5 = 97.5$ volts. The beam appears quite accurately at this voltage. It also conforms over the range through which

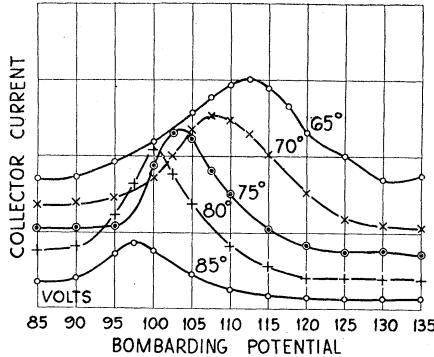


Fig. 16. Collector current vs. bombarding potential showing plane grating beams near grazing in $\{110\}$ -azimuth.

it can be followed to its appropriate grating formula, $\lambda = d_2 \sin \theta$, or $V^{1/2} \sin \theta = (97.5)^{1/2} = 9.88$. Values of $V^{1/2} \sin \theta$ taken from Fig. 16 are given in the last section of Table II.

These beams, as has been mentioned, are extremely sensitive to the presence of gas on the surface of the target. They fall off in intensity as the

¹⁰ In Fig. 15 the maxima near 50 volts in the curves for 65 and 70 degrees are due to the "54 volt" beam in early stages of its development. The curves in Fig. 15 should be compared with those in Fig. 9. The latter are for the target covered with gas; the "plane grating" beam is altogether lacking here, and the "54 volt" beam shows much more weakly than in the curves in Fig. 15. The rapid rise of the current in all colatitudes below 30 volts in Fig. 15 is not significant. It is due to a rapid increase of current in the incident beam which resulted, under the conditions of these measurements, from a focussing action within the electron gun.

gas collects, and disappear when the amount of gas on the surface is insufficient to affect more than slightly the intensity of beams occurring at higher voltages and smaller colatitude angles.

This behavior is consistent with the view already proposed that near grazing the intensity of scattering is determined almost entirely by the atoms in the topmost layer. When the crystal is clean this is a layer of regularly arranged nickel atoms which gives rise to the plane grating beams—when gas has collected on the surface it is a layer of gas atoms or molecules which may or may not give rise to diffraction effects of its own, but which, in either case, serves to absorb the radiation from the first layer of nickel atoms.

We thought that since the radiation in the grazing beams is scattered mostly by top layer atoms, the electrons in these beams would be more nearly homogeneous (made up more completely of full speed electrons) than those in beams at higher voltages and lower colatitude angles—beams more dependent for their intensities upon radiation scattered from atoms in lower layers. This appears, however, not to be the case. Current-voltage curves similar to those in Fig. 7 have been constructed for the grazing beams, and these indicate a distribution of speeds quite like that inferred from Curve III of Fig. 7.

We have looked also for the first order grazing beams from the plane gratings of third largest spacing, but have failed to find them. There should be twelve such beams (six in each of two new azimuths) appearing at 227 volts. It is to be expected, of course, that the higher voltage beams of this type will be weak on account of a less rapid extinction of the equivalent radiation in the metal. Here, as elsewhere, it appears a dependable procedure to infer characteristics of the equivalent radiation from known properties of electrons; if high speed electrons are less rapidly absorbed than low speed electrons, we may infer that short wave equivalent radiation is less rapidly absorbed than long.

We have looked (also without success) for other beams of this type grazing the inclined {111}-facets. The calculated positions of most of these beams fall so close to intense space lattice beams that it seems unlikely they could be found, even if present. On the other hand a careful exploration for one particular beam of this sort, the predicted position of which is not too close to any space lattice beam, was a complete failure.

SPACE LATTICE BEAMS

The diffraction pattern for radiation of a given wave-length scattered by a single atom layer is made up, as has already been described, of the plane grating beams from all of the gratings that can be constructed from lines of atoms in the layer. The beams occur in planes normal to the lines of these gratings, and satisfy the plane grating formula $n\lambda = d \sin \theta$, where d represents the constant of the particular grating considered. This relation is represented graphically by the straight lines in Fig. 17, in which wavelength λ is plotted against $\sin \theta$, for beams occurring in the principal azimuths. For a given value of λ the positions of the beams in the various orders may be read off from these lines.

We consider now what diffraction effects are to be expected when the incident waves are scattered not by a single layer of atoms, but by a number of such layers piled one above the other. We see that if the various atom layers are similarly oriented, the lines in Fig. 17 represent the wave-length angle relation for the diffraction beams from each layer in the pile, and that, therefore, they must represent the same relation for the beams issuing from

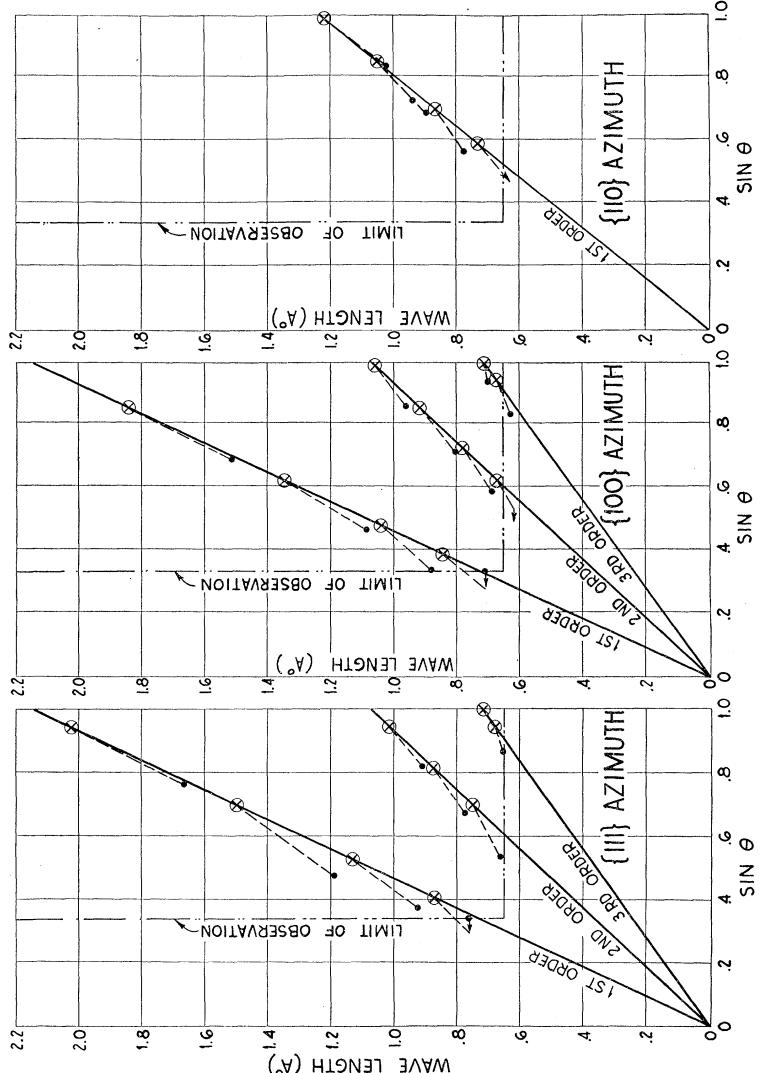


Fig. 17. Plots of the grating formulas in the three principal azimuths. Positions of theoretical x-ray beams are indicated by crossed circles and positions of actual electron beams by dots.

the pile of layers; the sum of the superposed beams is still the same set of beams. As far, therefore, as the positions of the beams are concerned the diffraction of waves by a pile of parallel and similarly oriented atom layers is the same as by a single layer.¹¹

¹¹ It is assumed in this paragraph that the atom plane is large enough to be highly resolving and that we are not interested in discrepancies of the order of the width of its diffraction beams.

There is, however, a great difference in the intensities of the beams in the two cases. In the case of scattering from a single layer the intensity of a given beam would be found ordinarily not to change greatly as the wave-length and position of the beam are varied. In the case of scattering from a pile of such layers, however, the intensity of the emergent beam is greatly affected by interference among the beams from the individual layers. If a great number of layers contribute individual beams of about the same intensity, as is the case in x-ray scattering, the intensity of the resultant beam will ordinarily be quite low. When, however, the beams from the individual layers emerge from the crystal in phase the intensity of the resultant beam passes through a strong maximum. The wave-lengths for which this occurs depend upon the way in which the atom layers are piled up—that is, upon the separation between successive layers and upon the lateral shift from one layer to the next. If the wave-length of the radiation within the crystal is not the same as outside this also will have an effect upon the occurrence of intensity maxima.

The purpose of setting forth these rather elementary matters at such great length is to bring out clearly that the intense space lattice beams are always also plane grating beams. If the wave-length and position of such a beam is represented by a point in Fig. 17 the point must fall somewhere on one of the plane grating lines.

If the incident beam in the present experiments were a beam of heterogeneous x-rays an array of Laue diffraction beams would issue from the crystal. The wave-lengths and positions of all such beams in the three principal azimuths within the range of our observations ($\theta > 20^\circ$, $\lambda > 0.63\text{\AA}$) are indicated in these figures by crossed cycles.

The plane grating lines for azimuths $\{111\}$ and $\{100\}$ are, of course, the same, but the occurrence of Laue beams in these azimuths is different. The wave-lengths that give rise to diffraction beams in the $\{111\}$ -azimuth are not the same, except in such orders as are exactly divisible by three, as those that give rise to beams in the $\{100\}$ -azimuth. And the same statement, *mutatis mutandis*, may be made, of course, in regard to the angles at which beams occur in the two azimuths. When the wave-lengths or angles of the beams of any order, not exactly divisible by three, are ordered according to magnitude the members belonging to one azimuth occur alternately with those belonging to the other. These characteristics result from the fact that to superpose the atoms of one layer upon those of an adjacent layer involves a displacement normal to the grating lines equal to $(2n+2/3)$ times the grating space— n representing any integer. If the odd fraction were one-half or zero, as in the $\{110\}$ -azimuth, these differences would not occur.

In this same figure we have indicated by dots the wave-lengths and positions of all sets of electron beams listed in Table I. The first thing to be noted about the dots is that they fall generally along the basic plane grating lines, and to this extent satisfy the fundamental requirement that a space grating beam shall be first of all a plane grating beam. It will be noted also that they occur along these lines at about the same intervals as the

crossed circles, and further that in the {111} and {100}-azimuths they exhibit the alternate occurrence characteristic of the crossed circles.

While the dots representing the sets of electron beams fall generally along the plane grating lines, one cannot fail to note that they actually fall off these lines—and systematically; they are above or to the left of the lines as one cares to view them. At the time of writing our note to "Nature" we believed that these departures could be accounted for by imperfections in the geometry of the apparatus. At present, with more data at our disposal, we are less certain that this is true. The fault of this explanation is that if the displacements of the dots from the lines result from imperfect alignment, angular displacement should be a function of angular position, and this condition seems not very well satisfied. The results of these investigations do not, however, rule out mechanical imperfection as the principal cause of these displacements, and for the present we shall assume it to be the only cause. We shall assume, that is, that the wave-lengths or voltages of the beams are correct but that the dots should be shifted to the right onto the lines. These shifts correspond to corrections in angle ranging from zero to about four degrees. We hope to remove the uncertainty here involved by measurements with a new tube now being constructed.

As the dots occur at about the same intervals along the lines as the crossed circles it is possible to associate each dot with a particular crossed circle—each set of electron beams, that is, with a particular set of x-ray beams, and consequently with a particular set of Bragg atom planes in the crystal. The associations which seem most natural are indicated by dotted lines in the figure. In the {110}-azimuth the association is imperfect as it appears that both the 0.893A and the 0.940A electron beams should be associated with the same x-ray beam. We have associated the stronger of these with the x-ray beam, and left the extremely weak electron beam at 0.940A without an apparent x-ray analogue. Also there is apparently a third order electron beam missing in the {111}-azimuth. We should expect this beam to be extremely weak.

We consider next the difference in position between each crossed circle and the location upon the line to which the associated dot is shifted as explained above. The positions of the lines are determined by the arrangement of atoms in a single plane and the scale factor of the structure, while the positions of the crossed circles are determined by the separation between successive planes and the lateral shift from one plane to the next. If the separation were decreased the crossed circles would all be moved downward along their lines; if it were increased they would be moved upward. Merely as a matter of description, therefore, we may say that a given dot when shifted occupies the position that its associated crossed circle would occupy if the separations between atom planes were decreased by a certain factor. This factor β may be calculated from the formula

$$\beta = \frac{\tan (\theta'/2)}{\tan (\theta_x/2)}$$

Here θ_x represents the colatitude angle of the Laue x-ray beam and θ' that of the associated electron beam after having been corrected to make the beam satisfy the grating formula exactly.

This factor β is found to be neither constant nor very regular in its behavior, but to increase generally with the speed of the electrons. The relation between the factor β and the bombarding potential is represented by the lower set of points in Fig. 18. There is a vague suggestion here that β approaches unity as a limiting value. If this is actually the case, it means, of course, that at sufficiently high voltages (short wave-lengths) there is no difference between the occurrence of x-ray and electron diffraction beams. This approach of the electron beam to the associated x-ray beam with

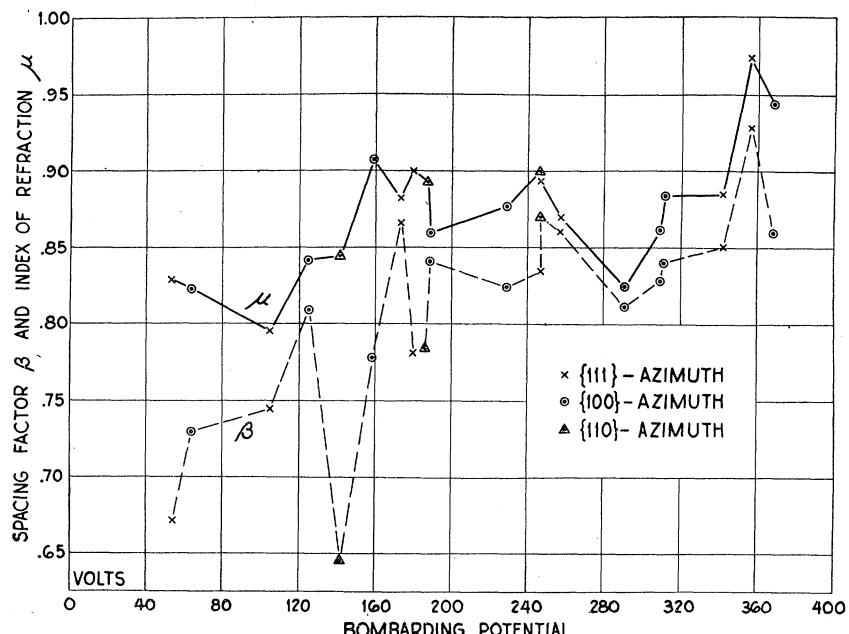


Fig. 18. Lower set of points—Spacing factor β vs. bombarding potential; Upper set of points—Index of Refraction μ vs. bombarding potential.

increasing voltage is shown very clearly by the series of three dots (Fig. 17) representing the first three orders of the beam whose first order is the 54 volt beam in the $\{111\}$ -azimuth ($\lambda = 1.67\text{\AA}$). The first, second and third order beams occur at $\theta = 50^\circ$, 55° and 62° respectively, while the corresponding x-ray beam occurs in each case at $\theta = 70^\circ$ ($\sin \theta = 0.94$).

It has been suggested by Eckart¹² that this β factor may be interpreted as an index of refraction of the nickel crystal for the equivalent radiation. If the index of refraction of a crystal for radiation of a given wave-length is other than unity, diffraction will indeed occur as if the spacing between crystal planes were altered by a certain factor. This factor is not, however,

¹² Eckart, Proc. Nat. Acad. Sci. **13**, 460 (1927).

equal to the index of refraction. For the case in which we are interested, that of normal incidence, and emergence from the same surface, the quantity appearing in the measurements as "spacing" factor β is given by

$$\mu = \frac{\beta}{\cos^2 \psi + \beta^2 \sin^2 \psi}$$

where ψ is the angle between the set of Bragg atom planes associated with the diffraction beam and the surface of the crystal. Values of μ calculated from this formula are represented by the upper set of points in Fig. 18. There is no greater regularity in the points representing μ than in those representing β .

There is the question, of course, as to whether the association of x-ray and electron beams indicated in Fig. 17 is the correct one. We might, for example, associate each dot in this figure with the next lower crossed circle instead of with the next higher, and there is at least one consideration favoring this procedure; the indices of refraction would then be slightly greater than unity for low speed electrons, and would approach unity for electrons of high speed. Such a dependence could be correlated with the increase in speed (decrease in wave-length) which an electron experiences on passing into a metal. In fact, we might naturally expect the index of refraction to be given by $\mu = [(V + \phi)/V]^{1/2}$, where ϕ represents the voltage equivalent of the work function of the metal.

The principal consideration opposed to this association is the absence of electron beams to be associated in the various orders with the x-ray beams of greatest wave-length. The absence of some of these might be accounted for by total reflection of the diffraction beam at the surface of the crystal. It seems unlikely, however, that the absence of analogues in three orders of the x-ray beams at $\theta = 70^\circ$ ($\sin \theta = 0.94$) in the $\{111\}$ -azimuth could be accounted for in this way. The absence of an electron beam to be associated with the first order x-ray beam of greatest wave-length in the $\{100\}$ -azimuth would seem decisively against this alternative association were it not that the strong grazing beam in this azimuth (Fig. 14) may, in fact, be this analogue. This matter of association should, we think, be regarded as still an open question to be decided by further and more precise measurements. For the present, however, we shall adhere to the associations of Fig. 17.

MISSING BEAMS

When electron and x-ray beams are associated as in Fig. 17 there is only one x-ray beam in the three principal azimuths for which an electron beam analogue predicted within the range of our observations has not been found. This is the third order beam in the $\{111\}$ -azimuth already mentioned. There are, however, three Laue beams in range in two new azimuths, one of wave-length 0.78A at $\theta = 74^\circ$ in the $\{331\}$ -azimuth, and two of wave-lengths 0.81A and 0.76A at $\theta = 86^\circ$ and $\theta = 68^\circ$ in the $\{210\}$ -azimuth. The first of these azimuths¹³ lies 11° from the $\{110\}$ -azimuth toward the nearest $\{111\}$ -

¹³ We have consistently designated each azimuth by the Miller indices of the densest plane of atoms the normal of which lies in the azimuth and in or above the surface of the crystal.

azimuth, and the second lies an equal interval on the opposite side of the $\{110\}$ -azimuth. A thorough but fruitless search has been made for the electron beam companions of these three Laue beams.

The total number of x-ray beams for which electron beam companions are expected within the range of our observations is twenty-four, and for twenty of these the electron beam companions have been found. The four sets of missing electron beams are all beams whose intensities we should expect to be low. They are all short wave-length beams lying not far above the plane of the target.

THIRD ORDER BEAMS IN THE $\{111\}$ AND $\{100\}$ -AZIMUTHS

The differences which have been noted between the occurrence of x-ray diffraction beams in the $\{111\}$ and $\{100\}$ -azimuths do not extend, as has been mentioned, to orders that are exactly divisible by three; the wavelengths and positions computed for the third order x-ray beams in one of these azimuths are identical with those computed for the third order beams in the other. Differences in intensity between corresponding beams might possibly result from a dependence of the scattering power of a single atom upon crystal direction, but, so far as we are aware, differences of this sort are not found in x-ray data.

It seemed reasonable to expect, therefore, that the third order electron beams in the $\{111\}$ -azimuth would be found identical in voltage, position, and possibly also in intensity, with beams of the same order in the $\{100\}$ -azimuth. This agreement, however, is not observed. Two pairs of beams are available for observation, and the beams of each pair have constants that are different. The data are given below in Table III.

TABLE III
Third order electron beams.

x-ray companion	$\{111\}$ -azimuth			$\{100\}$ -azimuth			
	θ	V	Int.	x-ray companion	θ	V	Int.
$\{55\bar{1}\}$	—	—	0.00	$\{711\}$	70°	310	0.15
$\{551\}$	60°	347	0.07	$\{822\}$	57	370	0.15

RESOLVING POWER OF THE CRYSTAL

It has been remarked (Fig. 10) that the space lattice beams are not sharply defined in voltage. The "54 volt" beam, for example, appears when the bombarding potential is about 40 volts and is still prominent in the curve for $V=64$ volts. In terms of equivalent wave-lengths the beam has appreciable intensity over a range $\Delta\lambda$ which is about one quarter as great as the equivalent wave-length of the beam at its maximum intensity. The corresponding ratios for beams of higher voltage are generally less, but even for beams for which V is greater than 300 volts values of $\Delta\lambda/\lambda$ are found as great as 0.07. We naturally conclude that as an optical instrument the crystal has low resolving power.

It might appear that this low resolving power is readily accounted for by the fewness of the atom planes contributing effectually to the scattering. In accounting for the "plane grating" beams we have already had occasion to assume that the extinction of the equivalent radiation in the metal is extremely rapid, and it may appear that no further assumption will be required to account for the low resolving power observed in the space lattice beams. It must be realized, however, that the resolving power of a space lattice is dependent not only upon the number of its atom planes, but also upon the number of grating lines in each plane, and that to ascribe the low resolving powers in the present case solely to the slight penetration of the radiation into the metal is to assume that the number of grating lines in each of the atom planes making up the lattice is relatively great—great compared with the number of effective planes. If the width of the lattice were the same as the width of the bombarded area this assumption would, of course, be amply justified. There are, however, at least two reasons for regarding the diffracting system not as a single lattice, but as a collection of very many extremely small lattices, similarly oriented but otherwise unrelated. A large part of the radiation proceeds, it would seem, from lattices not more than five or ten atom lines in width.

The first reason for ascribing this small width to the unit lattice is that on no other assumption have we been able to account for the great width of such peaks as those shown in Figs. 9, 14, 15 and 16. The width of these peaks is due in part, of course, to the width of the diffraction beam and to that of the collector opening, but these geometrical considerations are quite insufficient for the purpose. The curves referred to seem to show clearly that the resolving power of the topmost atom layer is quite low, and that consequently most of the radiation from this layer comes from independent gratings containing only a few atom lines each.

The second reason has to do with the relation between the voltage and the angular position of a space lattice beam in the various stages of its development. We have seen (Table II) that as a "plane grating" beam rises from the surface of the crystal the product $V^{1/2} \sin \theta$ remains constant. At one time we expected that the same relation would be found to obtain within a space lattice beam—that as the voltage was increased the beam would in all cases move upward to keep $V^{1/2} \sin \theta$ constant. This relation does in fact hold reasonably well for the "54 volt" beam. It holds less well, however, for the "65 volt" beam shown in Fig. 10, and fails entirely for most beams occurring at higher voltage. In many cases the total angular displacement during the growth and decay of the beam is not more than one-tenth that required by the plane grating formula. It may be shown, however, that the plane grating formula ($V^{1/2} \sin \theta = \text{constant}$) will describe the motion of the beam if the lattice is sufficiently wide, but that it will fail to do so otherwise. If the number of lines in the plane grating is not great compared with the number of atom planes, the plane grating relation will not obtain (except at the maximum of a beam—when the scattering from successive layers is exactly in phase) and departures from it will be in the direction of those that we have observed.

There are thus these two reasons for thinking that most of the scattered radiation proceeds from lattices not more than five or ten grating lines in width, and that the low resolving power of the plane gratings that make up the representative lattice is partly responsible for the low resolving power apparent in the space lattice beams.¹⁴ It seems probable that the independent lattices are to be identified with the individual facets of the principal set—those parallel to the general plane of the target.

If the resolving power of the crystal were determined solely by the rate of extinction of the radiation in the metal, it would be possible to make use of the intensity-voltage relation within a given beam to calculate a coefficient of extinction of the metal for radiation of the wave-length of the beam, as suggested by Eckart (*loc. cit.*), or to estimate the number of atom planes effective in the scattering, as suggested by Patterson.¹⁵ It may even be possible under the actual conditions of the scattering to evaluate these constants by making use also of the observed relation between the angular position of the beam and voltage. But the data which we have at present seem to us too inaccurate to make such calculations worth while. We hope to obtain data sufficiently precise for this purpose in the near future.

We have, however, calculated a rate of extinction from the data obtained for the "54 volt" beam with the surface of the target free from gas (not from the data of Fig. 10). As has already been mentioned, the product $V^{1/2} \sin \theta$ is very nearly constant in this beam, which means that its intensity-voltage relation is determined almost entirely by the rate of extinction. As the result of this calculation we have obtained the value 0.4 for the fraction by which the intensity of a beam of 54 volt equivalent radiation is reduced when such a beam passes normally through a single {111}-layer of nickel atoms. In terms of electrons this means presumably that when a 54 volt electron is incident normally upon such a layer the probability of its passing through without appreciable deflection or loss of energy is 0.6.¹⁶

INTENSITIES OF "PLANE GRATING" BEAMS

We return now to a further consideration of the intensity difference between the first order "plane grating" beams in the {100} and {111}-azimuths (Figs. 14 and 15). If these were truly plane grating beams no difference of intensity should be observed. We have already pointed out, however, that it is only in the limit, as θ approaches $\pi/2$, that these beams are due to scattering from a single atom layer. They are actually space lattice beams, and their occurrence and behavior are described by the same mathematical formulas that describe the Laue type of beam. The intensity of a "plane grating" beam depends upon the efficiency with which a single

¹⁴ It was shown in an earlier section that the apparent angular width of a space lattice beam (voltage constant) has more than double its least possible value as calculated from the dimensions of the apparatus. It seems probable from the present considerations that a considerable part of this additional width is to be ascribed to the low resolving power of the crystal.

¹⁵ A. L. Patterson, *Nature* **120**, 46, (1927).

¹⁶ This question of resolving power has been considered recently by F. Zwicky (*Proc. Nat. Acad. Sci.* **13**, 519 (1927)) in terms of the wave mechanics as formulated by Schrödinger and Born.

atom scatters the radiation incident upon it, and upon the rate of extinction of the radiation in the crystal. Such beams are not found when x-rays are scattered by a crystal because in this case both of these quantities are small.

The problem of finding which of the two beams under consideration should be the stronger resolves itself into finding in which azimuth the first order diffraction beams from successive atom layers are more nearly in phase near grazing emergence; the beam in this azimuth will be the more intense. These phase differences between beams from successive atom layers are readily deduced from the geometry of the lattice. Thus it may be shown that if the diffraction occurs as though the crystal were contracted normally to its surface to a fraction β of its normal spacing, this phase difference in the $\{111\}$ -azimuth will be given by

$$\alpha_{111} = \frac{4\pi n}{3} \left[\frac{\beta 2^{1/2} (1 + \cos \theta)}{\sin \theta} + 1 \right],$$

and in the $\{100\}$ -azimuth by

$$\alpha_{100} = \frac{4\pi n}{3} \left[\frac{\beta 2^{1/2} (1 + \cos \theta)}{\sin \theta} - 1 \right]$$

where n represents the order of the beam. Phase differences for $\theta = 75^\circ$ and for various values of β have been computed from these formulas, and are given in Table IV.

TABLE IV
First order phase differences for $\theta = 75^\circ$

Spacing Factor β	0.5	0.6	0.7	0.8
(Phase Diff) ₁₁₁	$2\pi + 1.8$	$2\pi + 2.5$	$4\pi - 3.0$	$4\pi - 2.2$
(Phase Diff) ₁₀₀	-0.3	0.4	1.2	2.0

From the results shown in Fig. 18 we expect that at 35 volts, the bombarding potential for these beams, the value of β will lie near 0.7. The phase difference in the $\{111\}$ -azimuth is, therefore, about 3.0 radians and that in the $\{100\}$ -azimuth about 1.2 radians. The beam in the $\{100\}$ -azimuth should therefore be the stronger, and this is what is actually observed.

THE TEMPERATURE EFFECT

It has been mentioned already that immediately after bombardment, while the target is still at a high temperature, the intensities of all beams, as far as observed, are low, and that as the target cools the intensities rise again to their normal values for room temperature. The behavior is illustrated by the curves in Fig. 19 in which the intensities of the beams at ($V = 54$ volts, $\theta = 50^\circ$) and ($V = 343$ volts, $\theta = 34^\circ$) in the $\{111\}$ -azimuth are plotted against time. The temperature of the target at zero time was perhaps 1000°K . The data for these curves were taken during the same run—observations on one beam being alternated with those on the other.

This temperature effect has not yet been studied in detail. There seems no reason for doubting, however, that it is the analogue of the Debye temperature effect observed in the diffraction of x-rays. This view is supported by the evidence of the curves in Fig. 19 that the higher voltage (shorter wavelength) beam is the more sensitive to temperature.

BEAMS DUE TO ADSORBED GAS

The "plane grating" beams and space lattice beams for which data have been given in previous sections are the only beams observed when the target is, as we believe, free from gas. When the target is not free from gas still other beams appear. Three sets of these have already been mentioned as the ones referred to in our note to "Nature" as anomalous; one set occurs in each of the three principal azimuths, and all attain their maximum intensity at or close to $\theta = 58^\circ$, $V = 110$ volts. These beams appear to depend for their existence upon the presence of a considerable amount of gas on the surface of the target; they were most intense before the first bombardment of the target, and were not found when the amount of adsorbed gas was known

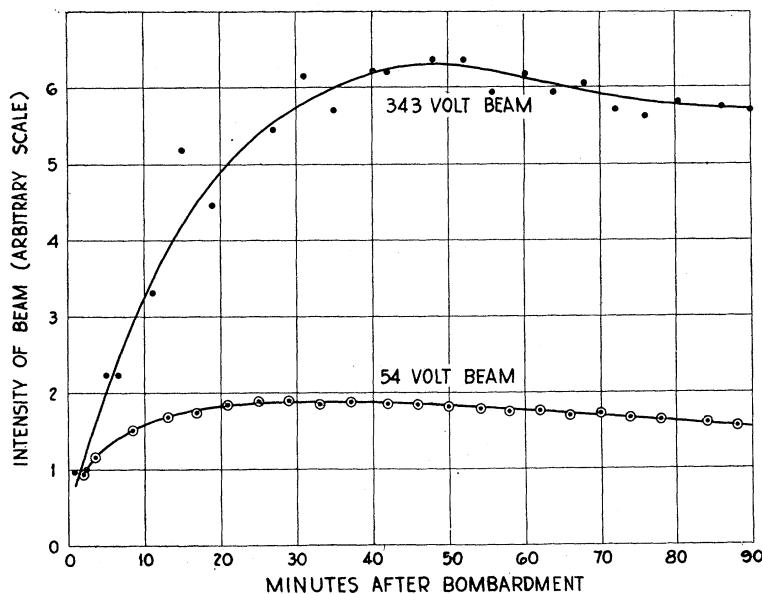


Fig. 19. Intensities of 54 volt beam and 343 volt beam vs. time, immediately after heating the target crystal.

to be slight. It should be possible to infer from the characteristics of these beams something about the arrangement of atoms to which they are due. It is clear, for example, that the arrangement is determined in part by the underlying nickel, since the beams occur in the principal azimuths of the nickel structure. We have been unable, however, to carry the deduction beyond this point.

With certain other beams due to gas we have been more successful. These constitute a family attaining greatest intensity when the quantity of adsorbed gas has a certain critical value. Various members of this family were observed first as beams of unusual behavior that appeared in places in which no beams were expected, and disappeared or changed in intensity for no apparent reason. Thus a beam was occasionally found in the {110}-azimuth at $\theta = 75^\circ$, $V = 25$ volts. No such beam could result from scattering

by the nickel crystal unless our entire theory is fallacious; the first order grazing beam for this azimuth appears at $V=97.5$ volts, and no diffraction beam is possible for any lower voltage.

A correlation was eventually established between the intensity of this 25 volt beam and that of the normal grazing beam in the same azimuth. It was found that the 25 volt beam was strongest when the intensity of the grazing beam had been reduced by adsorbed gas to about one-fifth of its maximum value, and that when the grazing beam was at its maximum intensity the 25 volt beam was not to be found. Furthermore the 25 volt beam was not found when the surface was so contaminated that the normal grazing beam was absent; nor was it present when the 110 volt "anomalous" beams could be found.

It was noticed also that the voltage of this beam is almost exactly one quarter the voltage of the normal first order beam occupying the same position. The equivalent wave-length is thus twice that of the corresponding

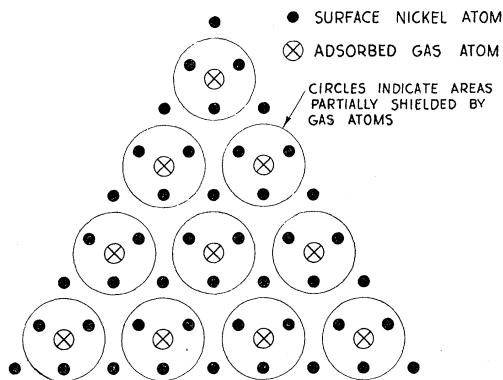


Fig. 20. Arrangement of gas atoms on the surface, and the topmost layer of nickel atoms.

first order beam, and the beam could be accounted for as radiation scattered by a layer of gas atoms of the same structure and orientation as the nickel atoms, but of twice the scale factor. Such a layer can be imagined built onto the surface layer of nickel atoms as indicated in Fig. 20. The first order diffraction beams from such a layer agree in wave-length and position with "one-half order beams" from the underlying lattice of nickel atoms, and as these latter have zero intensity the resultant beams should be those due to the single layer of gas atoms only. The beams should, therefore, move continuously upward as the voltage is increased without marked change in intensity. What is observed is not quite so simple; the beam can indeed be followed continuously from $\theta=85^\circ$, $V=24$ volts up to $\theta=30^\circ$, $V=98$ volts, but its intensity passes through broad maxima at $\theta=75^\circ$, $V=25$ volts, and at $\theta=45^\circ$, $V=50$ volts.

A possible explanation of this behavior may be pointed out. It will be noticed (Fig. 20) that the nickel atoms in the surface are of two sorts—those that are adjacent to gas atoms and those that are not. One-fourth of the

nickel atoms are of this latter class, and they together form a layer of the same structure, orientation and scale factor as the gas atoms. If the two classes of nickel atoms scatter radiation in different amounts—if, for example, the gas atoms shield the adjacent nickel atoms more effectually than the non-adjacent—then, of course, the layer of nickel atoms as a whole will give rise to a differential diffraction beam capable of interference with that due to the gas layer, and broad maxima of the sort observed will result.

It was anticipated, of course, that beams of the same nature would be found in the {111} and {100}-azimuths. These should appear at grazing at $V=32.5/4=8.1$ volts and should move upward with increasing voltage—reaching $\theta=20^\circ$ at $V=70$ volts. Observations cannot be made for bombarding potentials as low as 8 volts. The beams have been picked up, however, in both azimuths at $\theta=60^\circ$, $V=12$ volts, and have been followed upward to $\theta=25^\circ$, $V=45$ volts. Broad maxima occur at $\theta=35^\circ$, $V=25$ volts, and at $\theta=55^\circ$, $V=13$ volts in the {100}-azimuth; and in the {111}-azimuth a broad maximum occurs at $\theta=55^\circ$, $V=14$ volts, and another is indicated in the neighborhood of $\theta=25^\circ$, $V=45$ volts. It should be possible from these data to calculate the separation, or at any rate the apparent separation, of the gas layer from the crystal—but this has not yet been attempted.

We have further observed that these beams cannot be made to appear when the temperature of the target is somewhat above that of the room; glowing the filament back of the target raises the temperature of the target sufficiently to eliminate them entirely, although under these conditions gas still collects on the target, and reduces the intensities of the grazing beams. The explanation of this behavior may be that the melting point of the two dimensional gas crystal is not far above room temperature. We have not yet observed whether the beams disappear sharply at a critical temperature.

FURTHER EXPERIMENTS WITH GAS

In a final series of experiments the effect was studied of introducing large amounts of gas into the tube. The liquid air was removed from the charcoal tube, and the behaviors of the "54 volt" beam and of one of the anomalous "110 volt" beams were observed as the charcoal was heated. The latter of these beams had not been observed for several weeks—not since its disappearance during the first heatings of the target.

The initial effect of increasing the gas pressure was to decrease the intensity of the "54 volt" beam. Its intensity was, however, greater than that represented by curve *A* in Fig. 13 until the charcoal temperature reached 350°C. At about this time the anomalous beam made its appearance. Maintaining the charcoal at 350°C the 54 volt beam decreased rapidly in intensity, vanishing entirely within a few minutes. In the meantime the intensity of the anomalous beam increased, reaching a maximum at about the time the 54 volt beam vanished, after which it too decreased and finally vanished. The gas pressure within the apparatus, as shown by ionization measurements, was perhaps 10^{-4} mm Hg at this time.

Heating was discontinued and liquid air was replaced on the charcoal. This caused the pressure to return to a very low value, but it did not bring

back either the 54 volt beam or the anomalous beam. Heating the target by electron bombardment did, however, bring back the 54 volt beam to about its maximum intensity (curve *B*, Fig. 13). At this time the anomalous beam was found to be absent. The 54 volt beam did not maintain its initial intensity but decreased rapidly, indicating that the vacuum condition of the tube as a whole had been greatly impaired by heating the charcoal. No further tests were made.

SUMMARY OF ELECTRON BEAMS

Thirty sets of electron beams in all have been observed for bombarding potentials below 370 volts. Eleven of these occur in the {111}-azimuth, twelve in the {100}-azimuth and seven in the {110}-azimuth.

Twenty of the sets have been associated with twenty sets of Laue beams that would issue from the same crystal if the incident beam were x-rays.

Three sets are accounted for as "plane grating" beams which result from a preponderance of top layer scattering at angles near grazing emergence.

Six sets are attributed to scattering by adsorbed gas on the surface of the crystal, and the structure of the gas film giving rise to three of these beams has been inferred.

Some explanation has thus been given for twenty-nine of the thirty sets of beams. The remaining set occurs at $\theta=46^\circ$ in the {110}-azimuth for $V=170$ volts, and is quite weak.

The explanations used in accounting for the observed beams require the occurrence of still other beams that have not been observed. The total of these missing beams is at least eight; four space lattice beams (one third order beam in the {111}-azimuth, one first order beam in the {210}-azimuth and two first order beams in the {331}-azimuth); and four "plane grating" beams (first order {210} and {331}-azimuths, and second order {111} and {100}-azimuths).

Discrepancies have also been noted between the characteristics of the third order space lattice beams in the {111}-azimuth and those of their companions in the {100}-azimuth.

The possibility of carrying through these investigations to their present stage has depended very largely upon the cooperation we have received from a number of our colleagues here in the laboratory. We are particularly indebted to Drs. H. D. Arnold and W. Wilson for the encouragement they have given us and for the benefit of their criticisms. We have had the benefit also, in technical matters, of discussions with Drs. L. W. McKeehan, K. K. Darro and R. M. Bozorth.

We are indebted to Mr. H. T. Reeve for producing for us single nickel crystals of appropriate size—to Mr. C. J. Calbick for assistance in making the observations and for contributing not a little to their interpretation—and to Mr. G. E. Reitter for the great care with which he constructed the special apparatus and for his many contributions to its design.

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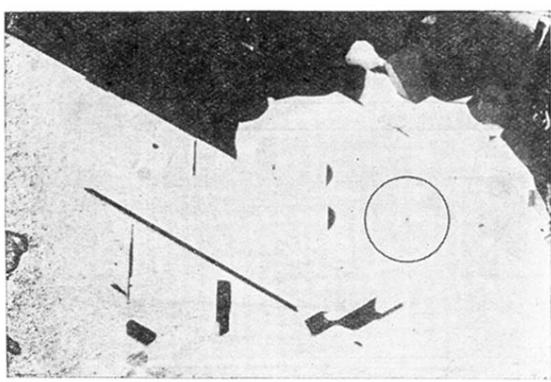


Fig. 5. Microphotograph of the nickel target.