X-RAY DIFFRACTION PATTERNS FROM PLANT FIBERS.

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Plate 3.

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The finer structure of the cell-wall of plant fibers was anticipated years ago in the theories of Nägeli, Strasburger, and others, but the actual existence of a lattice arrangement of structural units was not definitely established until x-ray methods were applied to the problem. Several years ago x-ray diffraction patterns were obtained from plant fibers and a space lattice was computed from the data thus made available. In an attempt, by the author, to confirm the conclusions of the earlier investigators, Herzog and Jancke, certain discrepancies led to an intensive study of the diffraction patterns. A different method was used and more than twenty new lines were found on the photographs, while some of those previously reported were missing. This lack of agreement reopened the field for further investigation. (Cf. Nature, 1925, cxvi, 243.)

Method and Apparatus.

The method of obtaining the data upon which this report is based was very similar to that of Hull for producing x-ray diffraction patterns from crystal powders, and the apparatus was practically the same as that described in many places in the literature. A quite detailed

1 Nägeli, K., Über den innern Bau der vegetabilischen Zellenmembranen, 1864.
2 Strasburger, E., Über den Bau und das Wachstum der Zellhauten, Jena, 1882, 225.
3 Herzog, R. O., and Jancke, W., Z. Physik, 1920, iii, 196.
5 Hull, A. W., Phys. Rev., 1917, x, 2nd series, 661; 1921, xvii, 2nd series, 571.

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description was given when reporting the work done on starch, which will furnish the reader with information concerning the principles of x-ray reflection involved in this work, and with the method of interpreting diffraction lines in terms of a space lattice.

A standard Coolidge x-ray tube with a water-cooled molybdenum anticathode was the source of radiation. It was run at 42,000 volts, and the exposures were usually 25 milliampere-hours. A zirconium oxide screen made the beam practically monochromatic with a wavelength of 0.71 Å, that of the Kα line. The cassette, or film holder, was much like that referred to in the earlier paper, with slight modifications made to allow for the rotation of a block of fibers. This block was built by laying thousands of fibers approximately parallel and compressing them into a tablet 3 mm. thick by 15 mm. wide, the length limited to that of the fibers. After squaring up the end of the tablet, a piece 3 mm. long was cut from it across the fibers, making a small block 3 × 3 × 15 mm. This small block (see Fig. 1) consisted of short lengths of fibers 3 mm. long, which were for the most part parallel to OC. The ends of the fibers appeared at the BOA face and at the face opposite.

The block, with an indicator needle attached to it, was placed at the center point of the cassette, where it was adjusted in such a way that when the needle was set at 0° on the protractor the OC edge of the block, and therefore most of the fibers, were parallel to the line of propagation of the x-ray beam. The beam, then, would pass lengthwise through most of the fibers. When the block was turned so that the needle pointed to 90°, the beam would pass through at right angles to the long axes of the fibers.

The diffraction patterns which were obtained from these two positions were quite different from each other. The pattern from the 0° position consisted of three prominent and three faint lines; that from the 90° position, of five or six strong lines and about as many weak ones.

Photographs were made from several kinds of fibers, but the lines obtained from ramie (Bommeria nivea) were used almost exclusively in this work because they were much more clean cut than those from either hemp (Cannabis sativa) or spruce (Picea sitchensis).

The lines on the photographs (Plate 3) indicate the existence of atoms or reflecting units of some kind arranged in sets of parallel planes, each line usually bearing a definite relation to a specific set of planes. The distance between the planes of a given set is computed from the formula \( n\lambda = 2d \sin \theta \), where \( \lambda \) is the wave-length of the x-rays used, \( d \) the distance between the planes, and \( \theta \) is the glancing angle determined from measurements of the lines on the photographs.\(^7\)

The accuracy with which the spacing, \( d \), could be determined was about 1 per cent. The blurring of some of the lines brought the error occasionally up to about 2 per cent, rarely more than that. For example: the 6.10 value was quite certain to \( \pm 0.07 \), 3.98 to about \( \pm 0.04 \), 2.58 to \( \pm 0.02 \), and 1.70 to \( \pm 0.01 \), the error becoming smaller with the smaller spacing values.

In the following paragraphs it is convenient to refer to the corresponding spacings, planes, and lines by using the same figures; thus, the planes which are spaced 3.98 Å. u. will be referred to as the 3.98 planes and the line they produce as the 3.98 line.

The photographs of some of the diffraction patterns are not readily reproduced in half-tone because considerable blackening is caused by a general scattering of the x-rays from certain positions of the block of fibers. From other positions the lines stand out clearly on the negative. The 90° position is an example of the latter while the 0° position is one from which the lines are blurred into the background between them (see Plate 3).

Each line was produced by a set of parallel planes in the wall of the fiber. In order to locate those planes with reference to some readily ascertained axis such as the long axis of the fiber, it is necessary to show that the orientation of the fibers in the block corresponds to the orientation of the planes which produced the lines. In other words, the data deal with the planes, while it is the fiber as a whole with which we have to work. These fibers (ramie) were only 0.07 mm. in diameter and very flexible since they were several cm. long. Since the length of the block was only 3 mm. along the OC edge it was obviously made up of pieces of these long fibers. In building up the block, absolute parallelism between the fibers was, of course, not obtainable; but by making a tablet with full length fibers first and then cutting the block (Fig. 1) from that, it is quite probable that most of the fibers were approximately parallel to one another and also to the OC edge of the block. The deviation of the fibers from true parallelism very probably corresponded to a fairly symmetrical frequency distribution. From a microscopic inspection of the block most of the fibers seemed to lie within a range of about 8° on each side of a mean. That is there were about as many fibers lying parallel to OC as there were at 2° to OC and approximately the same number at any given position up to about 8°, but rather suddenly the number dropped off so that a much smaller number were lying 10° to OC, only a few 15°, and scarcely any were found which were lying 20° to the OC edge of the block.

**Diffraction Patterns from the 0° Position.**

Returning now to the diffraction patterns, we find that when the indicator needle was set at 0° the photograph showed that 6.10, 5.40, and 3.98 planes were in proper positions to reflect their corresponding lines; when it was set at 2° the same lines appeared and their density was about the same as before; and when set at 4°, 6°, and 8° respec-
tively there was still very little if any difference in the density. At
10°, however, the lines were slightly less dense; at 15°, decidedly less
dense; at 20° they were scarcely visible and at 25° there was no trace
of them, for the short exposures given in taking this series of photo-
graphs. If now we compare these changes in density with the changes
in the number of fibers throughout the same range of angles, as given
in the paragraph above, we find a correspondence which indicates
that the planes responsible for the diffraction lines were parallel to
the long axis of the fiber.

There is another point which should be considered briefly here
before our conclusions may be fully justified. It concerns the glancing
angle of the various planes. It is evident from the preceding para-
graphs that the block of fibers could not act as a single large crystal,
and that at any given angle between the 0° and the 20° positions there
must occur a group of fibers in which planes, for example the 6.10,
would be oriented properly for reflection of their line. The number of
active fibers varied inversely with the angle, but not in direct proportion
variation. If our assumption is correct that the long axis and the
planes were parallel, then with the block set at 0° the 6.10 line would
be produced by a group of fibers which were lying 3° to the OC
line, since that is the glancing angle for the 6.10 planes; the 5.40 line would
be produced by another group of fibers which were lying about 4°,
and the 3.98 line by still another group slightly over 5° to the OC line.
Theoretically there would be more fibers in position to reflect the 6.10
line than the 3.98, and further there would be more fibers active in
reflecting the 6.10 line at the 0° position than, for example, at 4°.
The difference in number would be small, relatively, in both cases,
and the density of the lines would not be noticeably affected at ordi-
nary exposures, but by careful adjustment of the exposures it was possi-
bable to show a direct correspondence between the diffraction line at
its densest and the fibers which were parallel to OC, with a greater
degree of accuracy than was brought out in the preceding paragraphs.
We were able to show that the 3.98 planes were parallel to the long
axis to within an experimental error of 2°; the 6.10 and 5.40, within
an error of 4°.

The correctness of the assumption that these three sets of planes
were all parallel to the long axis of the fiber then seems highly probable,
and if this is so they must bear very definite relations to one another. These relations are brought out graphically in Fig. 2. By trial it was found that when 6.05 planes form a 90° angle with 5.35 planes, the diagonals, as shown by the solid diagonal lines in Fig. 2, are spaced 4.00. In the figure the planes are represented as perpendicular to the paper. Their spacings agree with the observed values within the limits of experimental error. Both observed and calculated values are presented in Table I for comparison.

The agreement seems to be sufficiently close to warrant our acceptance of one view of the elementary cell as a rectangle 6.10 Å u. by 5.40 Å u.

TABLE I.

Interplanar Spacing. 0° Position of Fiber Block.

<table>
<thead>
<tr>
<th>Observed</th>
<th>Calculated</th>
<th>Density of lines</th>
</tr>
</thead>
<tbody>
<tr>
<td>Å u.</td>
<td>Å u.</td>
<td></td>
</tr>
<tr>
<td>6.10</td>
<td>6.05</td>
<td>Strong.</td>
</tr>
<tr>
<td>5.40</td>
<td>5.35</td>
<td>Medium.</td>
</tr>
<tr>
<td>3.98</td>
<td>4.00</td>
<td>Very strong.</td>
</tr>
<tr>
<td>2.65</td>
<td>2.65</td>
<td>Weak.</td>
</tr>
<tr>
<td>1.98</td>
<td>2.00</td>
<td>Very weak, second order.</td>
</tr>
<tr>
<td>1.93</td>
<td>1.90</td>
<td>“ “</td>
</tr>
</tbody>
</table>

The orientation of these rectangles on a cross-section of a fiber is suggested by data from two sources. The fiber is a minute hollow cylindrical tube with walls about 0.02 mm. thick. The work of Strasburger\textsuperscript{8} and of W. L. Balls\textsuperscript{9} has demonstrated that the cellulose substance is deposited by the protoplasm of the cell, layer upon layer, on the inside of the wall, forming more or less concentric cylinders. If we assume the 6.10 dimension to be the distance between these concentric cylinders, there would be about 40,000 of them in a fiber whose wall is 0.02 mm. thick. The 5.40 dimension would be the distance between radial planes. In the diagram in Figs. 3 and 4 a cross-section

\textsuperscript{8} Strasburger, E., Über den Bau und das Wachstum der Zellhauten, Jena, 1882, Plate 3, Fig. 22.

of a fiber is represented. The wall is MN in thickness. The several sets of rectangles represent one view of as many groups of elementary cells, enormously out of proportion to the rest of the diagram. In them the 6.10 planes are tangential to the fiber, the 5.40 planes are radial. The long axis of the fiber and the planes are perpendicular to the paper. If one imagines a flat beam of x-rays to pass lengthwise through this cylinder, at an angle of 3° to the long axis, it will be seen that there are only two regions, AA, from which the 6.10 planes will reflect to produce a 6.10 line. If now the angle is changed from 3° to about 4°, that is to the glancing angle for the 5.40 planes, a 5.40 line would be reflected from two—and only two—other regions, BB,
tions. It seems more probable that on the cross-section view of a fiber, as in Fig. 3, a group of atoms is centered around each intersection, since the atoms we are dealing with are about 1.5 Å.u. in diameter or less. Also, it seems quite probable that the group is not arranged around the intersection in a radially symmetrical manner since the 6.10 line is denser than the 5.40, but that more atoms are associated with the 6.10 plane than with the latter. The effect of that arrangement may perhaps be more clearly pointed out in Fig. 4 where the conventional elliptical characters, centered at each intersection of the lines representing 6.10 and 5.40 planes, indicate oblong groups of atoms. Planes such as OD or OE would be quite likely to fail to produce diffraction lines because of the annulling effect of interleaved planes.

**TABLE II.**

<table>
<thead>
<tr>
<th>From Fig. 4.</th>
<th>Calculated.</th>
<th>Observed.</th>
<th>Density.</th>
</tr>
</thead>
<tbody>
<tr>
<td>OA</td>
<td>6.05</td>
<td>6.10</td>
<td>Strong.</td>
</tr>
<tr>
<td></td>
<td>5.35</td>
<td>5.40</td>
<td>Medium.</td>
</tr>
<tr>
<td>OB</td>
<td>4.00</td>
<td>3.98</td>
<td>Very strong.</td>
</tr>
<tr>
<td>OC</td>
<td>2.65</td>
<td>2.65</td>
<td>Weak.</td>
</tr>
<tr>
<td>OD</td>
<td>1.90</td>
<td>1.93</td>
<td>“</td>
</tr>
<tr>
<td>OE</td>
<td>2.46</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>OA</td>
<td>1.73</td>
<td>1.98</td>
<td>Second order.</td>
</tr>
</tbody>
</table>

Those represented by OB or OC might produce faint lines, if any. In Table II the observed and calculated values are included for comparison with the lines predicted from Fig. 4. The agreement makes the assumption seem worth further consideration when determining the lattice structure.

By way of summary we may say that the data up to this point seem to justify the conclusions that the structural units in the fiber are arranged in concentric layers around the long axis, and that in those layers they are located so that they occur also in radial planes. There seems to be no way of determining whether it is the 6.10 or the 5.40 value that is associated with the concentric layers. The use of 6.10 for that spacing, in the discussion, was merely accidental. There was neither experimental nor theoretical basis for the choice.
Diffraction Patterns from the 90° Position.

When the block of fibers was turned so that the indicator needle pointed to 90°, a diffraction pattern was obtained which consisted of about a dozen lines. With the block in that position most of the fibers were oriented approximately at right angles to the path of the beam of x-rays. Any lines, therefore, which appeared on the photographic film were necessarily reflected from planes which were approximately at right angles to the long axis of the fiber. The more exact position of those planes with respect to the long axis and to the planes already considered was determined by the same method as that used at the 0° position. Five photographs were taken, with short exposures, at intervals of 2° on each side of the 90° position. From them it was possible to establish rather satisfactorily several things. The angles formed by the 2.58 planes with the 6.10, 5.40, and 3.98 planes, and therefore with the long axis of the fiber, were found to be 90° ± 2°. Four other sets of planes, although not so readily located because their lines are weaker and not so sharply marked, seemed to be parallel also to the 2.58 planes. Another set of photographs of longer exposures brought out three more very faint lines whose planes seemed to be parallel to the 2.58 planes. In all, there were eight sets of planes apparently parallel to one another and perpendicular to the long axis of the fiber. In Table III are given the interplanar spacing values for these transverse planes, and the density of the lines which they produced.

TABLE III.

Diffraction Lines from 90° Position of Fiber Block.

<table>
<thead>
<tr>
<th>Interplanar spacing, Å</th>
<th>Density of line</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.15</td>
<td>Strong.</td>
</tr>
<tr>
<td>3.40</td>
<td>Medium.</td>
</tr>
<tr>
<td>2.58</td>
<td>Very strong.</td>
</tr>
<tr>
<td>2.03</td>
<td>Strong.</td>
</tr>
<tr>
<td>1.70</td>
<td>Very weak.</td>
</tr>
<tr>
<td>1.46</td>
<td>&quot; &quot;</td>
</tr>
<tr>
<td>1.29</td>
<td>Weak.</td>
</tr>
<tr>
<td>1.14</td>
<td></td>
</tr>
</tbody>
</table>
The 2.03 line seemed to be composed of several superimposed lines, which have not been satisfactorily resolved. On some of the negatives the value was computed as 2.01, on others 2.04, depending upon the position of the block.

The four other prominent lines were readily measurable within the limits of experimental error. The 2.58 line might be considered as the second order reflection of 5.15 planes, except for the very obvious discrepancy in the densities. It seems more probable that there were planes interleaved half-way between the 5.15 planes, which were not equal to the latter in reflecting power, analogous to the situation in rock salt in which the (111) planes are alternately weak and strong reflecting planes. The result produced there, just as here between the 2.58 and the 5.15 is a weak first and strong so called second order reflection. A similar situation seemed to exist between the 3.40 and the 1.70 planes. The relationship which seemed to exist between the various members of this group of parallel planes will be brought out later when the lattice structure is developed.

In Plate 3 it will be seen that planes spaced 2.58 and 1.70 form prominent lines which are too dense, as explained above, to have been produced by reflections of higher orders. The atoms of which the planes were composed are about 1.30 Å.u, and 1.50 Å.u. in diameter. It would seem fairly probable, then, that in a lengthwise direction of the fiber the atoms would be less segregated into groups than they seem to be in a tangential or radial transverse direction. In other words, from a cross-section view of a fiber the structural units would appear as fairly well isolated groups of atoms, while on a longitudinal view of the fiber they would appear separated laterally, but more closely associated in a lengthwise direction, forming fairly continuous strings of atoms running lengthwise of the fiber.

**Diffraction Patterns from Positions between 0° and 90°.**

With structural unit groups so arranged that they lie in transverse planes and at the same time form longitudinal layers in the fiber, they must also form diagonal planes which would produce diffraction lines when the block of fibers is turned to various positions between 0° and 90°.

This prediction was very clearly verified by several series of photo-
graphs taken with the block set at 5° intervals in some cases and 10° intervals in others, between 0° and 90°. A bewildering number of lines was obtained.

When sorted out, each line was found to appear from several adjacent positions and usually the position of greatest density was readily determined. For example the 4.35 line appeared on the negatives from the 50°, 60°, and 70° positions. The rate of change in density indicated that its densest position was probably between 60° and 65°. A series of short exposures taken at 2° intervals showed the position of most intense reflection to be very close to 62°.

### TABLE IV.

**Diffraction Lines from Positions between 0° and 90°.**

<table>
<thead>
<tr>
<th>Position of fiber block</th>
<th>Interplanar spacing and density of lines.</th>
</tr>
</thead>
<tbody>
<tr>
<td>80°</td>
<td>2.01 m.; 1.94 vvw.; 1.69 w.; 1.44 vw.</td>
</tr>
<tr>
<td>70°</td>
<td>3.20 s.; 1.94 vvw.; 2.35 w.; 1.25 vw.; 1.10 vw.</td>
</tr>
<tr>
<td>60°</td>
<td>4.35 vs.; 2.17 s.; 2.35 w.; 1.82 vvw.; 2.97 s.; 2.62 s.; 1.10 w.</td>
</tr>
<tr>
<td>50°</td>
<td>2.62 vs.; 2.17 s.; 1.82 vvw.; 3.95 vw.</td>
</tr>
<tr>
<td>40°</td>
<td>6.40 vvw.; 3.10 vw.; 1.88 vw.; 3.95 vw.; 1.55 vw.</td>
</tr>
<tr>
<td>30°</td>
<td>2.93 vvw.; 3.10 vw.; 1.88 vw.; 2.65 w.</td>
</tr>
<tr>
<td>20°</td>
<td>2.65 w.</td>
</tr>
<tr>
<td>10°</td>
<td>2.65 w.; 2.34 vvw.</td>
</tr>
</tbody>
</table>

**vs., very strong; s., strong; m., medium; w., weak; vvw., very weak; vww., only a faint trace.**

In Table IV are given the interplanar spacing, density of line, and position of the fiber block at which the line appeared at its densest, for all of the lines obtained between the 0° and 90° positions. No attempt was made to estimate the position closer than the interval reading, which in this case is 10°. Wherever a line seems to be equally dense in two adjacent positions it is recorded in two places.

By revolving the block on an OA axis (Fig. 1) every possible set of planes was brought into an effective position for producing a diffraction line. That becomes evident only when one recalls that the block does not act as a single large crystal, and that the thousands of cylindrical fibers are laid more or less parallel to one another and to the OC edge of the block. In order to verify this, other blocks were made with
the fibers running parallel to $OA$ instead of $OC$, and as expected the only lines produced were those which had appeared in the former when set at $0^\circ$.

**SUMMARY.**

The rather long discussion just given seemed necessary in order to establish certain points before attempting to develop the lattice structure and before working out the identity of the structural unit of the ramie fiber.

1. Certain planes, 6.10, 5.40, 3.98, etc., as given in Table I, run lengthwise of the fiber; that is, they are parallel to the long axis.

2. These planes are in agreement with the assumption that one set, either the 6.10 or the 5.40 is tangential to the fiber and forms concentric cylinders, with the long axis of the fiber as the long axis of the cylinders; the other set, either the 5.40 or the 6.10 cuts the former at right angles and therefore its planes are radial with respect to the fiber, theoretically all of them meeting at the long axis, as indicated in the cross-section of a fiber in Fig. 3.

3. Other planes, 5.15, 3.40, 2.58, etc., as given in Table III, are transverse planes which form right angles with the long axis and therefore with the planes of Table I.

4. All of the planes are composed of reflecting units, probably groups of atoms, located at the intersections of the planes. This being the case, other reflecting planes must occur at other angles to the long axis. This prediction is verified by the lines given in Table IV.

5. The structural units in the wall of the fiber thus form a space lattice, the elementary cell of which is an orthorhombic structure.

6. Comparatively little can be said as yet concerning the structural unit. The unit is very probably composed of a group of atoms which are more or less closely packed together. If the groups were visible they would appear, in a cross-section of a fiber, as closely packed groups of atoms, 6.10 Å.u. from center to center of groups in one direction, and 5.40 Å.u. at right angles to that. In a longitudinal section, however, they would appear less compact and might even lose the appearance of groups in forming long strings of atoms which would extend lengthwise of the fiber.
By establishing the positions of the planes in the wall of the fiber, as in Tables I, III, and IV, it would seem that all dimensions of the elementary cell, and the size and character of the structural unit, could be determined. Work along these lines is now in progress.

EXPLANATION OF PLATE 3.

Fig. 1. X-ray diffraction patterns of ramie fibers. Figures opposite the lines indicate the spacing of the planes which produced them. Those at the bottom give the position of the indicator needle at which the photograph was taken.