

five, convergence was attained.

The agreement of calculated with observed structure amplitudes shows that the results are at least approximately correct. For the 429 observable data, the discrepancy factor is 0.16. Of the 436 data (not included among those systematically absent) below threshold, 388 have  $|F_{\text{calc}}|$  below the minimum observable  $|F_{\text{obs}}|$  and 48 have  $|F_{\text{calc}}|$  greater than threshold values, in most cases, only slightly greater. For all the reflections systematically absent, the  $|F_{\text{calc}}|$  was less than the minimum observable value. This overall agreement is satisfactory considering the nature of the crystals and the necessary idealization of the structure.

The structure of the helical molecule was given only one variable, namely, the radius,  $r$ , in the least-squares analysis. Because the final value of  $r$ , 0.95 Å, is very near that given by Prins et al.<sup>7</sup> the structure of the molecule remains very much like the one they described: The S-S distance is 2.07 Å and the S-S-S bond angle, 106°; the shortest non-bonded S-S distance is 3.30 Å and the dihedral angle  $S_1S_2S_3 - S_2S_3S_4$  is 95°.

None of the large number of intermolecular distances (Fig. 3) is significantly shorter than 3.37 Å, the shortest intermolecular distance

previously reported for sulfur.

We have indicated earlier that the pressure-induced fibrous modification of sulfur is identical with  $\psi$ -sulfur. The rotation photograph about the fiber axis of a crystal of the pressure-induced phase exactly matches that of a fiber pattern of  $\psi$ -sulfur. The literature<sup>12</sup> contains erroneous conclusions regarding the indexing of this pattern. For completeness, we give here (Table 1) our indexing based on the pseudo-orthorhombic cell and confirm that the data obtained by Tuinstra from the fiber pattern of  $\psi$ -sulfur agree with those from the pressure-induced phase. There is greater resolution in our data, however. (We hope to have a somewhat more detailed discussion of this indexing published elsewhere).

The crystals of the fibrous phase of sulfur have remained intact (at atmospheric pressure and room temperature), that is, with no apparent transformation to any other phase, for a period of approximately 3½ years.

With respect to phase (I), we have proposed that it is probable that the sulfur helical molecules lie in planes perpendicular to a 32.4 Å repeat distance as in the fibrous phase, but because it is lamellar, the helices are skew to each other in alternating planes. Phase I has

Table 1. Indexing of Rotation Photograph of  $\Psi$ -Sulfur

| Tuinstra |       |       |              | Present Work |       |       |              | Tuinstra |       |       |              | Present Work |       |       |              | Tuinstra |       |       |              | Present Work |       |       |              |  |  |
|----------|-------|-------|--------------|--------------|-------|-------|--------------|----------|-------|-------|--------------|--------------|-------|-------|--------------|----------|-------|-------|--------------|--------------|-------|-------|--------------|--|--|
| $Q_o$    | $Q_o$ | $Q_c$ | $h\ k\ \ell$ | $Q_o$        | $Q_o$ | $Q_c$ | $h\ k\ \ell$ | $Q_o$    | $Q_o$ | $Q_c$ | $h\ k\ \ell$ | $Q_o$        | $Q_o$ | $Q_c$ | $h\ k\ \ell$ | $Q_o$    | $Q_o$ | $Q_c$ | $h\ k\ \ell$ | $Q_o$        | $Q_o$ | $Q_c$ | $h\ k\ \ell$ |  |  |
| 473      | 478   | 467   | 002          | *            | 2625  | 2634  | 2,12,3       |          |       |       |              | 3792         | 3774  | 3759  | 4,14,3       |          |       |       |              |              |       |       |              |  |  |
|          |       | 610   | 080          | *            | 3253  | 3284  | 245          |          |       |       |              |              | 3826  | 3800  | 425          |          |       |       |              |              |       |       |              |  |  |
| 613      | 613   | 620   | 042          | *            | 3713  | 3701  | 2,16,3       |          |       |       |              | 4093         | 4036  | 4043  | 4,18,1       |          |       |       |              |              |       |       |              |  |  |
| 1842     | 1847  | 1839  | 0,12,2       | *            | 4092  | 4137  | 2,20,1       |          |       |       |              | 4751         | 4719  | 4715  | 4,10,5       |          |       |       |              |              |       |       |              |  |  |
| 2050     | 2046  | 2022  | 044          | *            | 4451  | 4501  | 2,12,5       |          |       |       |              | *            | 5639  | 5629  | 4,14,5       |          |       |       |              |              |       |       |              |  |  |
| 2431     | 2422  | 2439  | 0,16,0       | *            | 5582  | 5570  | 2,16,5       |          |       |       |              | *            | 6472  | 6503  | 4,22,3       |          |       |       |              |              |       |       |              |  |  |
| 2470     | 2475  | 2480  | 084          | *            | 5839  | 5814  | 2,24,1       |          |       |       |              | *            | 6957  | 6910  | 467          |          |       |       |              |              |       |       |              |  |  |
| 2868     | 2894  | 2906  | 0,16,2       | *            | 6099  | 6089  | 247          |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |
| 3244     | 3234  | 3242  | 0,12,4       | *            | 6610  | 6547  | 287          |          |       |       |              | 2250         | 2230  | 2233  | 660          |          |       |       |              |              |       |       |              |  |  |
| 4265     | 4271  | 4208  | 006          |              |       |       |              |          |       |       |              | 2379         | 2372  | 2396  | 622          |          |       |       |              |              |       |       |              |  |  |
|          |       | 4278  | 0,20,2       | 946          | 950   | 939   | 370          |          |       |       |              | 2707         | 2706  | 2701  | 662          |          |       |       |              |              |       |       |              |  |  |
| 4316     | 4345  | 4309  | 0,16,4       | *            | 1032  | 1026  | 332          |          |       |       |              | 2800         | 2823  | 2843  | 6,10,0       |          |       |       |              |              |       |       |              |  |  |
|          |       | 4360  | 046          |              | 1165  | 1160  | 1178         | 352      |       |       |              | 3284         | 3300  | 3311  | 6,10,2       |          |       |       |              |              |       |       |              |  |  |
| 4845     | 4843  | 4817  | 086          | 1165         | 1160  | 1178  | 352          |          |       |       | 3797         | 3807         | 3798  | 624   |              |          |       |       |              |              |       |       |              |  |  |
| 5449     | 5455  | 5487  | 0,24,0       | 1222         | 1224  | 1244  | 390          |          |       |       |              | 4172         | 4089  | 4103  | 664          |          |       |       |              |              |       |       |              |  |  |
|          |       | 5579  | 0,12,6       | 1414         | 1414  | 1407  | 372          |          |       |       |              | *            | 4202  | 4225  | 6,14,2       |          |       |       |              |              |       |       |              |  |  |
| 5618     | 5619  | 5680  | 0,20,4       | 1691         | 1706  | 1712  | 392          |          |       |       |              | *            | 4762  | 4712  | 6,10,4       |          |       |       |              |              |       |       |              |  |  |
| *        | 6631  | 6646  | 0,16,6       | 2104         | 2107  | 2093  | 3,11,2       |          |       |       |              | *            | 4923  | 4977  | 6,18,0       |          |       |       |              |              |       |       |              |  |  |
|          |       |       |              |              | 2377  | 2352  | 314          |          |       |       |              | *            | 5484  | 5444  | 6,18,2       |          |       |       |              |              |       |       |              |  |  |
|          |       |       |              |              | 2431  | 2428  | 334          |          |       |       |              | *            | 5640  | 5628  | 6,14,4       |          |       |       |              |              |       |       |              |  |  |
| 962      | 967   | 941   | 191          |              | 2532  | 2550  | 3,13,2       |          |       |       |              | *            | 6171  | 6136  | 626          |          |       |       |              |              |       |       |              |  |  |
| 1146     | 1133  | 1113  | 113          | 2500         | 2532  | 2550  | 3,13,2       |          |       |       |              | *            | 6521  | 6501  | 6,22,0       |          |       |       |              |              |       |       |              |  |  |
| 1300     | 1318  | 1322  | 1,11,1       |              | 2578  | 2581  | 354          |          |       |       |              | *            | 6952  | 6968  | 6,22,2       |          |       |       |              |              |       |       |              |  |  |
| 1361     | 1380  | 1342  | 153          |              |       | 2616  | 3,15,0       |          |       |       |              | *            | 7070  | 7050  | 6,10,6       |          |       |       |              |              |       |       |              |  |  |
| 1933     | 1934  | 1876  | 193          | 3110         | 3099  | 3083  | 3,15,2       |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |
| 2250     | 2251  | 2257  | 1,11,3       |              |       | 3114  | 394          |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |
| 2978     | 2948  | 2922  | 1,17,1       | *            | 3171  | 3226  | 3,17,0       |          |       |       |              | 2732         | **    | 2699  | 711          |          |       |       |              |              |       |       |              |  |  |
| 3113     | 3086  | 3060  | 135          | 3517         | 3489  | 3495  | 3,11,4       |          |       |       |              | 2808         | **    | 2776  | 731          |          |       |       |              |              |       |       |              |  |  |
|          |       | 3212  | 155          | *            | 5547  | 5512  | 3,23,0       |          |       |       |              | 2950         | **    | 2928  | 751          |          |       |       |              |              |       |       |              |  |  |
| 3255     | 3233  | 3248  | 1,15,3       | *            | 6295  | 6290  | 3,13,6       |          |       |       |              | 3470         | **    | 3461  | 791          |          |       |       |              |              |       |       |              |  |  |
| 3470     | 3454  | 3441  | 175          | *            |       | 7885  | 3,27,2       |          |       |       |              | 3712         | **    | 3710  | 733          |          |       |       |              |              |       |       |              |  |  |
| 3560     | 3548  | 3608  | 1,19,1       | *            | 7916  | 7962  | 318          |          |       |       |              | 3867         | **    | 3843  | 7,11,1       |          |       |       |              |              |       |       |              |  |  |
| 3790     | 3750  | 3746  | 195          | *            | 8263  | 8296  | 3,25,4       |          |       |       |              |              | **    | 3863  | 753          |          |       |       |              |              |       |       |              |  |  |
| 3860     | 3875  | 3857  | 1,17,3       | *            | 8856  | 8881  | 3,21,6       |          |       |       |              | 4136         | **    | 4092  | 773          |          |       |       |              |              |       |       |              |  |  |
| 5130     | 5135  | 5118  | 1,15,5       | *            | 9270  | 9287  | 3,27,4       |          |       |       |              | 4337         | **    | 4397  | 793          |          |       |       |              |              |       |       |              |  |  |
|          |       | 5789  | 117          |              |       |       |              |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |
| 5900     | 5858  | 5865  | 137          | 1318         | 1316  | 1300  | 461          |          |       |       |              | 5414         | **    | 5406  | 10,2,1       |          |       |       |              |              |       |       |              |  |  |
|          |       | 6123  | 1,25,1       |              |       |       |              |          |       |       |              | 5550         | **    | 5520  | 10,4,1       |          |       |       |              |              |       |       |              |  |  |
|          | 6096  | 6144  | 1,23,3       | 1942         | 1936  | 1910  | 4,10,1       |          |       |       |              | 6017         | **    | 5978  | 10,8,1       |          |       |       |              |              |       |       |              |  |  |
| 6200     | 6321  | 6246  | 177          | 2250         | 2253  | 2235  | 463          |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |
|          |       | 6932  | 1,11,7       | 2866         | 2856  | 2824  | 4,14,1       |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |
| *        | 7005  | 7058  | 1,25,3       |              |       | 2844  | 4,10,3       |          |       |       |              |              |       |       |              |          |       |       |              |              |       |       |              |  |  |

\* Not reported by Tuinstra.

\*\* Not measured in present work.