Conclusion

From the orientation of the water molecules in $MgSO_4.4H_2O$, as revealed by the neutron diffraction study, it can be concluded: (1) that the hydrogen bonds in a salt hydrate can be more bent than was known from previous investigations, (2) that a hydrogen atom of a water molecule need not necessarily participate in a hydrogen bond, (3) that the orientation of the water molecule in a salt hydrate is not only determined by the hydrogen bond geometry, but by the electrostatic forces between it and the cation as well.

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Kikuchi Pattern from a Silicon Wedge

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Under special conditions of illumination at 80 keV, the Kikuchi pattern of a thin single-crystal silicon wedge shows a dark cross within the intersection of the 110 and 101 dark zones. The pattern is interpreted as a combination of Bragg reflection effects, acting on the continuum of electrons produced by inelastic scattering.

In a series of extinction-contour studies on silicon single crystals, a dark Kikuchi figure has been repeatedly observed, which does not lend itself to interpretation as a simple defect line or band. Special conditions of thickness and illumination are required to emphasize the effect. It is similar to phenomena described qualitatively for other materials by Moliere (1961) and Selme (1963).

The silicon sample is a flake, of approximately one micron thickness, cut from a dislocation-free silicon single crystal* and finished by mechanical polishing, then etched, according to the methods of Dash (1956, 1958, 1959). The orientation is normal to a [001] axis. The flake has been mechanically broken, so as to provide cleavage surfaces, some of which form irregular wedges with the front or back faces; in these wedges a considerable degree of transmission is obtained at 80 keV.

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* The author is indebted to the late W. C. Dash, of the General Electric Research Laboratory, for this specimen.

The Kikuchi patterns are observed in a JEM 6A electron microscope, with the condenser beam concentrated in a circular spot about 7 microns in diameter, slightly overlapping the edge of the wedge. No field limiting aperture is used; the small size of the illumination spot defines the area producing the diffraction pattern. Under these conditions the Laue spot pattern from the thin part of the wedge is superimposed on the Kikuchi pattern from the thicker region immediately adjoining, as may be seen in Fig. 1. The intermediate lens of the microscope is focused for optimum clarity in the Kikuchi pattern. This is a positive print from the microscope plate; *i.e.* light areas correspond to high intensity.

The Kikuchi figure in question is the strong dark cross to the lower left of the marker spot indicating the direction of the primary beam. It occurs within the intersection of the 100, 010, 110, and 101 dark zones (Fig. 2), where no Kikuchi line due to a simple Bragg reflection is expected. The cross appears in strong contrast only when the crystal is illuminated at the corner of the 110 and 100 low intensity zones, as in Fig. 1. Under this condition of illumination, there is a strong 'pendellösung' effect in more than one Bragg reflection simultaneously, giving rise to thickness-contour fringes of high contrast, in the image of the crystal wedge. As many as seven 110 fringes are regularly observed at 80 keV. Beam 1 (Fig. 2) contains the dominant 110 reflection. Beams 2 and 3 may be thought to derive from 1 and *P*, respectively, by the 101 Bragg reflection, which is somewhat weaker here because the center of *P* lies somewhat above the 101 Bragg angle. The 100 Bragg reflection, from *P* to 2, is apparently weaker than these two dominant ones.

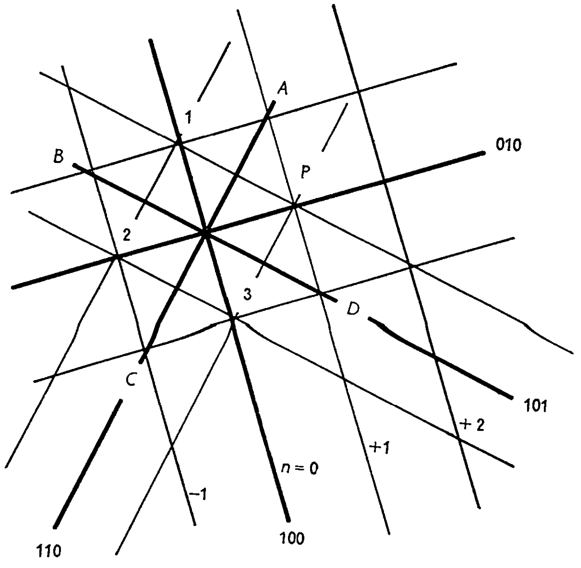

Fig. 2. Diagram of the Kikuchi pattern in Fig. 1. *P*: direction of primary beam.

The cross figure is aligned with the axes of the 100 and 010 dark zones. If the diffraction diagram is visualized as a distribution of transverse momentum, among the electrons emerging from the rear face of the crystal, the electrons which appear on these axes have their momentum aligned parallel to the (100) or (010) planes, which are densely packed and widely spaced. Thus the dark figure might be interpreted as a selective absorption of the type predicted by two-beam 'anomalous transmission' theory of Hirsch, Howie, Hashimoto & Whelan (1960). Such an interpretation has, in fact, been proposed by P. Selme (1963).

In this connection it should be noted that silicon has been shown by Borrmann, Hartwig & Irmler (1958) to provide a strong 'anomalous transmission' effect for X-rays, and by Kato & Lang (1959) to show attenuated 'pendellösung' fringes in X-ray diffraction. The conditions of illumination of Fig. 1 (strong simultaneous 110 and 101 reflections) would favor a strong 'anomalous absorption' at lattice sites along the 001 rows, due to inner-shell ionization and bremsstrahlung processes.

However, a different interpretation can be advanced, which involves no mechanism of selective absorption and follows the conventional interpretation of Kikuchi patterns as developed by Shinohara (1932) and Kambe & Miyake (1954). It should be noted that when the cross figure appears, it is always accompanied by a clear Kikuchi pattern of high contrast, which may be seen extending to the edges of Fig. 1. The inelastic continuum, originating around the diffraction beams *P*, 1, 2, 3 extends out to the intersections *A*, *B*, *C*, *D*, producing bright crosses at these points which are similar in shape and width to the dark central figure. These crosses lie at the intersection of the [100] and [010] zone edges, and they suggest a simpler mechanism to explain the dark central figure.

Consider the continuum lying above *P* and 1, outside the [101] zone, but inside *A*. This continuum lies close enough to the edge *P*–1 to be Bragg-reflected into the strip inside 2 and 3, by the 101 reflection, as the electrons pass through the last extinction length of the crystal before emerging from the rear face, Those electrons in this continuum which lie along the [100] zone edge *P*–*A* or the [010] zone edge 1–*A* will tend to be Bragg-reflected across these zones, however, giving rise to the bright lines at 2–*B* and 3–*D*. Thus there will be defect lines in the region inside 2 and 3, corresponding to the preference of these electrons for the 100 or 010 reflection, to the 101 reflection.

Thus the cross may be produced by simultaneous Bragg reflections of unequal strength, acting on the continuum of electrons produced by inelastic scattering. Although inelastic scattering is definitely involved in this interpretation, it need not be of the 'selective absorption' type assumed by Hashimoto, Howie & Whelan.

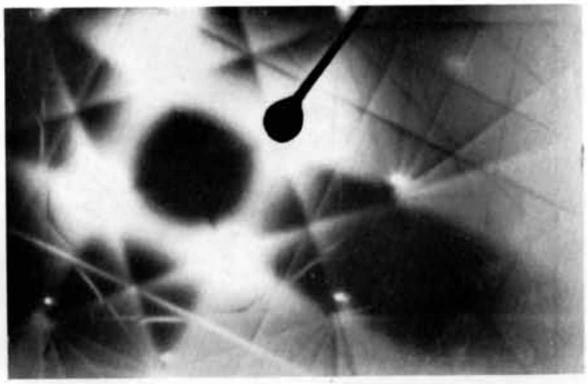

Fig. 1. Kikuchi diffraction pattern of a thin single-crystal silicon wedge, at 80 keV. Diffracting area is limited by the size of the illumination spot.

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A weaker effect of the same general type can sometimes be seen along the diagonals *A–C*, *B–D*, outside the square *P*123. In this case the lines of the cross are narrow, and the contrast considerably weaker. This pattern is probably due to competition between the first-order 010 and the second-order 101 Bragg reflections, with the latter producing a narrow defect line in the broad strip illuminated by the former.

This interpretation of the effect is in accord with the conventional interpretation of Kikuchi patterns, as developed by von Laue (1935). Shinohara (1932), and Kambe & Miyake (1954). Recent Kikuchi-line observations in silicon by Raether and collaborators (Dimigen, 1961; Hietel & Meyerhoff, 1961; Raether, 1962; Kunz, 1961; Creuzberg, 1963) have indicated that a plasmon energy-loss process produces the inelastic angular continuum which gives rise to the Kikuchi lines. The plasmon energy-loss is more readily reconciled with this conventional theory of Kikuchi patterns than with a 'selective absorption' theory of diffraction. The recoil momentum of the plasmon in studies of single (Kunz, 1961; Creuzberg, 1963; Boersch, Miessner & Raith, 1962; Geiger, 1962) and plural (Marton, Simpson, Fowler & Swanson, 1962) scattering has shown close agreement with a free-electron-gas model; hence it is not expected to exhibit a strong Fourier component at the lattice position in reciprocal space, which would give a 'selective absorption' analogous to the X-ray Borrmann effect.

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Note added in proof. — The orders of 100 identified in Fig. 2 as −1, +1, +2 are in fact the −2, +2, +4 orders of simple cubic 100. Odd orders are suppressed throughout the azimuth by the face-centered character of the diamond lattice, except at some regions near the [110] and [103] axes, where 'umweganregung' effects appear.

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