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Scattering of X-rays by Defect Structures. III. The Effect of Interstitial Atoms and Vacancies

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The effect, on the X-ray diffraction pattern, of a random distribution of interstitial atoms and vacancies in a single crystal of copper has been calculated on the basis of a model proposed by Huntingdon for the displacement of the atoms round such defects. Interstitials have a much greater influence than vacancies, and should produce observable X-ray effects when their concentration exceeds about 1%. Iso-diffusion contours near reciprocal-lattice points have a very characteristic shape.

1. Introduction

The irradiation of certain crystals with fast particles introduces lattice defects which are thought to be approximately equal concentrations of interstitial atoms and vacancies. The distribution of these defects throughout the crystal can be inferred only rather indirectly, but it appears that in some cases it can be regarded as a mixture of isolated interstitials and vacancies, together with regions of more concentrated damage. These conclusions have come mainly from resistivity measurements on irradiated metals, and optical measurements on alkali halides. Tucker & Senio (1955*a*, 1955*b*) have shown experimentally that pronounced X-ray scattering effects of the type predicted by Huang (1947) result from a sufficient concentration of such localized static lattice defects. In this paper we calculate the intensity of diffuse scattering, and the change of intensity of the Laue–Bragg reflexions that would result from a random distribution of interstitials and vacancies. We also consider briefly the effects of other possible distributions of defects. The effects predicted are qualitatively the same as were predicted by Huang, since we have used essentially the same model to represent the displacements of atoms by a defect, but we have been able to put them on a quantitative basis for a particular material, namely copper. The choice of material was dictated by the fact that for copper the displacement of atoms around a vacancy or an interstitial has been calculated by an approximate method (Huntingdon, 1953). This choice is unfortunate in that comparison of theory with experiment would be very difficult, as Cooper, Koehler & Marx (1954) have shown that appreciable thermal recovery of the metal occurs even at temperatures as low as 40° K. Our method of calculation, and possibly some of the conclusions, should, however, apply to other irradiated materials of simple crystal structure, including diamond, which is known to show X-ray effects at room temperature (Tucker & Senio, 1955*b*).

2. X-ray scattering by a single defect

In this section we make use of the theory given by Cochran (1956; referred to here as Part I) to calculate the effect of a single defect. A single interstitial atom is imagined, occupying the body-centre position of a face-centred cubic unit cell and at the centre of a spherical crystal of radius $\mathscr{R}$. The displacement $\boldsymbol{\varepsilon}^1$ of surrounding atoms at $\mathbf{r}$ can, unless r is small, be idealized as that due to a centre of pressure in a continuous elastic medium, which is (Huang, 1947; Eshelby, 1954)

$$\boldsymbol{\varepsilon}^1 = c\left\{\frac{\mathbf{r}}{r^3}+\frac{2(1-2\sigma)}{1+\sigma}\,\frac{\mathbf{r}}{\mathscr{R}^3}\right\}. \qquad (2\cdot1)$$

Here σ is Poisson's ratio, and, putting $\gamma = 3(1-\sigma)/(1+\sigma)$, the displacement of an atom at the surface of the sphere is

$$\varepsilon^1_{\text{max.}} = \gamma c/\mathscr{R}^2\,, \qquad (2\cdot2)$$

from which it can be shown that the fractional change in average unit cell side is

$$\frac{da}{a} = \frac{4\pi\gamma c}{3Nv}\,, \qquad (2\cdot3)$$

where $v = \frac{1}{4}a^3$ is the atomic volume, and N the number of atoms in the crystal. The theory given in Part I shows that the displacement which is effective for X-ray scattering is that given by (2·1), less any component that merely contributes to an expansion of the lattice. From (2·2), this component is $\gamma c\mathbf{r}/\mathscr{R}^3$, and the effective displacement is

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^1-\frac{\gamma c\mathbf{r}}{\mathscr{R}^3} = c\left(\frac{\mathbf{r}}{r^3}-\frac{\mathbf{r}}{\mathscr{R}^3}\right). \qquad (2\cdot4)$$

We have verified that the term in $\mathscr{R}^{-3}$ has a negligible effect on the intensity, and shall therefore take $\boldsymbol{\varepsilon} = c\mathbf{r}/r^3$.

For copper the value of c has been calculated as

$1{\cdot}8_7 \pm 0{\cdot}3_7$ (Huntingdon, quoted by Tucker & Sampson (1954)). The expression (2·1) cannot apply to atoms which are close to the interstitial, and for the 38 atoms within a sphere of radius $r_0 = 4{\cdot}7$ Å we have taken the displacements to be radially outwards and of amount shown in Fig. 1. These displacements are as given by Huntingdon's (1953) calculations, except that we have taken second nearest neighbours to be displaced as shown.

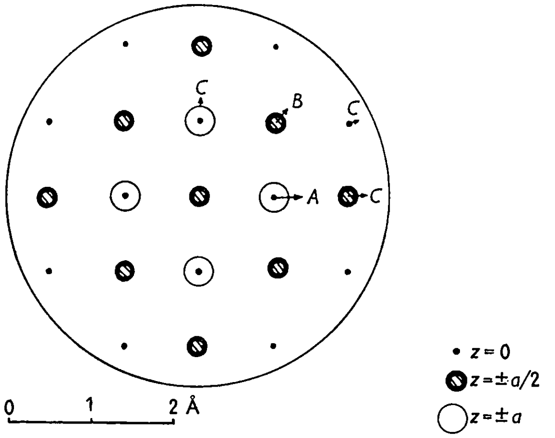

Fig. 1. Representation of displacements around the interstitial atom, assumed to be at the centre of a sphere of radius 4·7 Å. Positions of neighbours denoted in projection along the cube axis, atoms at different levels being shown in different sizes. Radial displacement for 6 nearest neighbours (A) is 0·39 Å, for 8 next nearest (B) is 0·13 Å and for the 24 third nearest neighbours is 0·07 Å.

The Fourier transform of a defect δ, as defined in Part I, is now given exactly by

$$T_\delta(\mathbf{S}) = f + f \sum_L (-\cos 2\pi \mathbf{R}_L\cdot\mathbf{S} + \cos 2\pi(\mathbf{R}_L + \boldsymbol{\varepsilon}_L)\cdot\mathbf{S}) , \tag{2·5}$$

where $\mathbf{R}_L$ locates an atom whose effective displacement is $\boldsymbol{\varepsilon}_L$, and $\mathbf{S}$ is a vector in reciprocal space. We may conveniently regard $T_\delta(\mathbf{S})$ as the sum of a contribution from atoms with $R_L < r_0$, denoted $T_{\delta 1}(\mathbf{S})$, and one from those with $r_0 < R_L < \mathscr{R}$, denoted $T_{\delta 2}(\mathbf{S})$. To a good approximation we then have (cf. equation (3·6) of Part I),

$$T_{\delta 2}(\mathbf{S}) = f \sum_L \left\{ -\tfrac{1}{2}\left(2\pi \frac{c}{R_L^3} \mathbf{R}_L\cdot\mathbf{S}\right)^2 \times \cos 2\pi \mathbf{R}_L\cdot\mathbf{S} - 2\pi \frac{c}{R_L^3} \mathbf{R}_L\cdot\mathbf{S} \sin 2\pi \mathbf{R}_L\cdot\mathbf{S} \right\} . \tag{2·6}$$

The sum is over all atoms for which $r_0 < R_L < \mathscr{R}$.

If we now take $S \leq 1{\cdot}3$ Å^{-1} (limit of Cu K radiation) and note that the maximum displacement of an atom beyond r_0 is 0·07 Å for $c = 1{\cdot}87$ Å^3 and $a = 3{\cdot}60$ Å, it is found that at least near reciprocal-lattice points the first term in the bracket of (2·6) is quite negligible compared with the second, except exactly *at* reciprocal-lattice points, where the second is zero. Reciprocal-lattice points are at $\mathbf{S} = \mathbf{H}$, where $\mathbf{H} = (\mathbf{h}+\mathbf{k}+\mathbf{l})a^*$ and $h+k$, $k+l$, and $h+l$ are even. We write $\mathbf{S}-\mathbf{H} = \mathbf{g}$, so that $\mathbf{g}$ is as defined by James (1948) in the theory of thermal diffuse scattering. It then follows from Part I that

$$T_{\delta 2}(\mathbf{S}) = -Sf \sum_{\mathbf{H}} (-1)^{h+k+l} \frac{4\pi c}{vg} \times \cos(\mathbf{S}, \mathbf{g}) \left\{ \frac{\sin 2\pi r_0 g}{2\pi r_0 g} - \frac{\sin 2\pi \mathscr{R} g}{2\pi \mathscr{R} g} \right\} . \tag{2·7}$$

The factor $(-1)^{h+k+l}$ appears because of the choice of origin. Except for $g \approx \mathscr{R}^{-1}$, the term $\sin 2\pi\mathscr{R}g/2\pi\mathscr{R}g$ can be ignored, and unless g is comparable with a^* ($= a^{-1}$) the summation in (2·7) can be adequately approximated by one term, especially as we have chosen a comparatively large value for r_0, so that near $\mathbf{S} = \mathbf{H}$, we have

$$T_{\delta 2}(\mathbf{S}) = -(-1)^{h+k+l} \frac{4\pi c S f}{vg} \cos(\mathbf{S}, \mathbf{g}) \frac{\sin 2\pi r_0 g}{2\pi r_0 g} . \tag{2·7a}$$

The transform of the remainder of the defect is given by

$$T_{\delta 1}(\mathbf{S}) = f + f \sum_L (-\cos 2\pi \mathbf{R}_L\cdot\mathbf{S} + \cos 2\pi(\mathbf{R}_L + \boldsymbol{\varepsilon}_L)\cdot\mathbf{S}) , \tag{2·8}$$

the summation being restricted to 38 atoms with $R_L < r_0$, and the first term, f, representing the contribution of the interstitial atom itself. This expression was evaluated numerically, with the help of the EDSAC, in the neighbourhood of the reciprocal-lattice points (2, 0, 0) and (4, 0, 0), in the section $l = 0$ of reciprocal space. The functions $(1/f)T_{\delta 1}(\mathbf{S})$ and $(1/f)T_{\delta 2}(\mathbf{S})$, along the line from $S = 0$ through (4,0,0) are shown in Fig. 2. It might be thought from the relative smallness of $T_{\delta 1}(\mathbf{S})$ on this line, that the atoms within 4·7 Å of the interstitial do not greatly

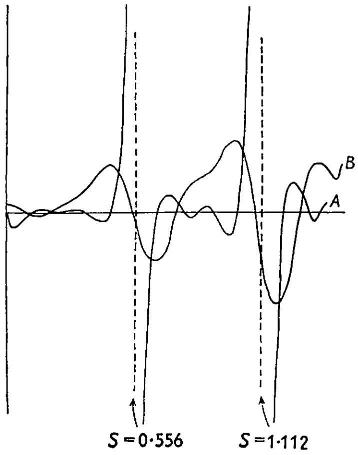

Fig. 2. Functions (A) $(1/f)T_{\delta 2}(\mathbf{S})$ and (B) $(1/f)T_{\delta 1}(\mathbf{S})$ along the line from the origin of the reciprocal lattice through (4, 0, 0). Broken vertical lines denote the reciprocal lattice points (2, 0, 0) and (4, 0, 0).

influence the diffuse intensity. That this is incorrect will be apparent on referring to Fig. 3, which shows the distribution $(1/f)T_{\delta 1}(\mathbf{S})$ in a section of reciprocal space. Only in the immediate neighbourhood—denoted by circles—of the reciprocal-lattice points is $(1/f)T_{\delta 2}(\mathbf{S})$ much the larger. Hence it is clear that the greater part of the total diffuse intensity, or volume integral of $|T_\delta(\mathbf{S})|^2$ over reciprocal space, originates from the displacement of near neighbours of the interstitial. It is, however, true that near reciprocal-lattice points, which are the only regions in which the diffuse intensity is relatively strong, (and for which our calculations are accurate), the contribution from the displacement of more remote atoms is the greater. It of course follows that the greater part of the reduction of Laue–Bragg intensity comes from the displacement of atoms within 4·7 Å of the interstitial. Indeed, a rough calculation shows that the displacement of nearest neighbours affects the Laue–Bragg intensity to about the same extent as the displacement of all atoms which are more than 4·7 Å away.

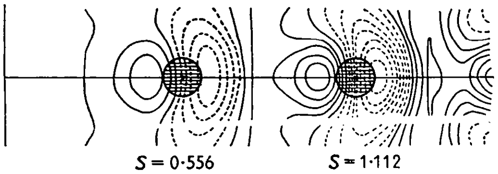

Fig. 3. Distribution of $T_{\delta 1}(\mathbf{S})$ in a section of reciprocal space. The three-dimensional distribution has almost cylindrical symmetry about the line from the origin through (4, 0, 0). The circles around the reciprocal-lattice points (2, 0, 0) and (4, 0, 0) mark the regions where $T_{\delta 2}(\mathbf{S})$ is very large.

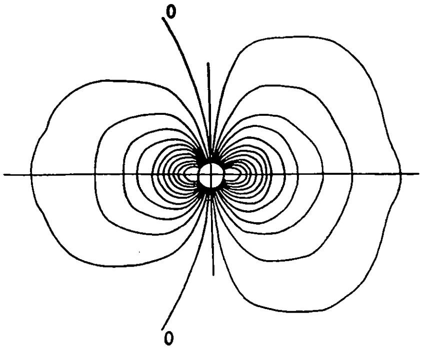

Fig. 4. Iso-diffusion contours around (2, 0, 0). The zero contour cuts the line through the origin and (2, 0, 0) at right angles at the reciprocal-lattice point, giving the typical lemniscate shape for the contours. Values of $(1/f)|T_\delta(\mathbf{S})|$ are shown; contours at 5, 10, 15, ...

From Part I we now have that the diffuse intensity is given by

$$J_2(\mathbf{S}) = |T_\delta(\mathbf{S})|^2 . \qquad (2{\cdot}9)$$

Iso-diffusion contours have been calculated in the neighbourhood of (2, 0, 0), and are shown in Fig. 4. Those near (4, 0, 0) would be similar, with the intensity increased by a factor 4, very nearly.

The change of intensity of a Laue–Bragg reflexion depends on $T_\delta(\mathbf{H}) = T_{\delta 1}(\mathbf{H}) + T_{\delta 2}(\mathbf{H})$. Values of $T_{\delta 1}(\mathbf{H})$ for $\mathbf{H} = 2\mathbf{a}^*$ and for $\mathbf{H} = 4\mathbf{a}^*$ were obtained in the course of the numerical calculation of (2·8). From (2·6), for these values of $\mathbf{H}$, we have

$$T_{\delta 2}(\mathbf{H}) = -\tfrac{1}{2} f \sum_L \left(\frac{2\pi c x_L H}{R_L^3}\right)^2 , \qquad (2{\cdot}10)$$

where x_L is the component of $\mathbf{R}_L$ in the direction of $\mathbf{H}$. Numerical evaluation gives $T_{\delta 2}(\mathbf{H}) = -5{\cdot}18H^2 f$. From Part I, the height of the Laue–Bragg peak, expressed as a fraction of that from the perfect crystal, is

$$\frac{J_1^1(\mathbf{H})}{J_1(\mathbf{H})} = 1 + 2\,\frac{T_\delta(\mathbf{H})}{F(\mathbf{H})} = 1 + 2\,\frac{T_\delta(\mathbf{H})}{Nf} . \qquad (2{\cdot}11)$$

As might have been expected, Laue–Bragg intensity decreases nearly in proportion to H^2. This 'artificial temperature factor' will not be isotropic in reciprocal space, but its value for any reciprocal-lattice point can be estimated from Table 1 to be about $1-18H^2/N$.

The calculation of the intensity effects which result from a vacancy could be made in the same way, with allowance for the fact that the vacancy is at (0, 0, 0) of the primitive trigonal unit cell, but since the intensity effects are nearly proportional to c^2, and the value of c for a vacancy is about one-fifth of that for an interstitial (Tucker & Sampson, 1954) it would be unrealistic to include a contribution from the vacancies when the value of c^2 for interstitials is uncertain by some 40%.

3. Scattering by randomly distributed defects

The results given in § 2 form the basis on which more realistic calculations can be made. We first of all use the theory given in Part I, which we would expect to apply when the concentration of defects is small, so that each atom is influenced almost entirely by a near defect, and the displacements are otherwise small. The interstitials are assumed to be randomly distributed, but this will include the case of a random distribution of paired vacancy–interstitials, since the influence of a vacancy is small. Putting the ratio of

Table 1. *Calculation of the change in Laue–Bragg intensity*

Position	H	$T_1(\mathbf{H})$	$T_2(\mathbf{H})$	$T(\mathbf{H})$	$T(\mathbf{H})/fH^2$	J_1^1/J_1
2, 0, 0	0·555	$-1{\cdot}12f$	$-1{\cdot}60f$	$-2{\cdot}72f$	−8·80	$1-5{\cdot}44/N$
4, 0, 0	1·111	$-5{\cdot}02f$	$-6{\cdot}40f$	$-11{\cdot}42f$	−9·25	$1-22{\cdot}84/N$
					(−9·0 average)	

the number of interstitial atoms to the total number of atoms equal to p, it follows from Part I that

$$J_2(\mathbf{S}) = Np|T_\delta(\mathbf{S})|^2 = p(V/v)|T_\delta(\mathbf{S})|^2 , \qquad (3\cdot1)$$

where V is the volume of the crystal, and near $(2,0,0)$ values of $|T_\delta(\mathbf{S})|^2$ are as given in Fig. 4. The reflexions $(2, 0, 0)$ and $(4, 0, 0)$ will be reduced by a factor $1-18\,pH^2$, which for $(4, 0, 0)$ and $p = 1\%$ is 0·78.

The general theory developed in Part II (Cochran & Kartha, 1956) gives identical results for small values of p, and not very different results even for $p = 3\%$, about the greatest concentration that can be expected (Kinchin & Pease, 1955). In applying the theory of Part II one finds that $T_{\delta 2}(\mathbf{S})$ is still given by (2·7), but $T_{\delta 1}(\mathbf{S})$ should now be taken as the first term in the expansion of (2·8) and the comparatively small effect of the interstitial itself cannot be allowed for, so that

$$T_{\delta 1}(\mathbf{S}) = -2\pi f \sum_L \boldsymbol{\varepsilon}_L \cdot \mathbf{S} \sin 2\pi \mathbf{R}_L \cdot \mathbf{S} \, . \qquad (3\cdot2)$$

We have evaluated this function along the line through $(4, 0, 0)$, and in Fig. 5 we compare the result with that given by (2·8). It will be seen that the general effect will be to give larger values for $|T_\delta(\mathbf{S})|$ than were given by the first theory. Near reciprocal-lattice points, however, where $T_\delta(\mathbf{S})$ is controlled by $T_{\delta 2}(\mathbf{S})$, the value of the former is not significantly changed from the value we derived earlier (Fig. 4). The diffuse intensity is now given by

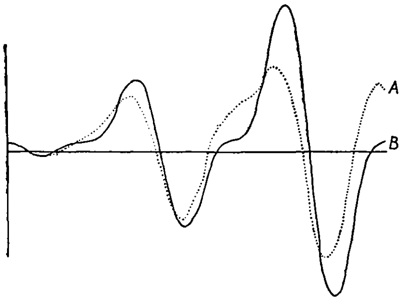

Fig. 5. Function $(1/f)T_{\delta 1}(\mathbf{S})$ along the line through the origin and $(4, 0, 0)$. The curve (A) gives the result of calculation using one expression and (B) that obtained with another appropriate to the extended theory.

$$J_2(\mathbf{S}) = p(V/v)e^{-2M}|T_\delta(\mathbf{S})|^2 , \qquad (3\cdot3)$$

where $M = 2\pi^2 pN\overline{(\boldsymbol{\varepsilon}\cdot\mathbf{S})^2}$. When $\mathbf{S}$ passes through $(4, 0, 0)$ it is readily calculated that $2M = 25\cdot6pS^2$. For $(4, 0, 0)$ and $p = 1\%$, $e^{-2M} = 0\cdot74$; for $p = 3\%$, $e^{-2M} = 0\cdot41$. Thus we may predict that as p increases, and conditions change from those for which our first treatment applies to those for which the second applies, the iso-diffusion contours near a reciprocal-lattice point remain approximately the same in shape, but the relative amount of diffuse intensity in this region decreases somewhat, to the gain of other regions in reciprocal space. This follows from the fact that the fractional change in Laue–Bragg intensity is about the same on both treatments, namely 0·78 and 0·74 for $p = 1\%$, 0·33 and 0·41 for $p = 3\%$ (for 4, 0, 0); and all the decrease must be distributed somewhere in reciprocal space. This agrees with the observation that on the whole a larger value of $|T_\delta(\mathbf{S})|$ is given by the second treatment.

It might be as well at this stage to emphasize the approximate nature of our calculations, and in particular to point out that Huntingdon's calculations of the effect of an interstitial, supplemented by the assumption of an inverse-square displacement of atoms at greater distances, cannot be entirely correct although they form the only basis for the calculation of X-ray scattering effects available at present.

4. Possible effects of a non-random distribution of defects

If the defects are not taken to be distributed at random, it is difficult to make quantitative predictions of the intensity. While the theory can be adapted, it seems unlikely when there is an agglomeration of interstitials, or of vacancies, that a principle of superposition will apply, and the following remarks should be regarded as having at best a qualitative significance. Suppose for example that Np interstitials are concentrated into Np^1 spheres each of radius R_0, the distribution of the p/p^1 interstitials being random inside each sphere and the agglomerates being randomly distributed in the crystal. The diffuse intensity can then be shown to be

$$Np(p/p^1)|T_\delta(\mathbf{S})|^2 Q^2(g)e^{-2M}, \quad \text{near} \quad \mathbf{S} = \mathbf{H} \, .$$

Here $Q(g)$ is the transform of a sphere of radius R_0, and therefore does not differ much from $\sin 2\pi R_0 g/2\pi R_0 g$. The effect, therefore, is to increase the diffuse intensity within roughly R_0^{-1} of reciprocal-lattice points, and to decrease it elsewhere. The characteristic 'lemniscate' shape of the iso-diffusion contours is, however, preserved, and (always of course assuming that the principle of superposition remains true) the lattice expansion and the Laue–Bragg intensities are the same as when the interstitials are distributed randomly throughout the crystal.

The 'displacement spikes' of Brinkman (1954), or 'thermal spikes' of Seitz (1949) cannot be regarded at all as regions in which the displacements of atoms can be described as the superposition of those caused by individual vacancies for interstitials. We might regard them as spherical regions in which atoms are so disordered as to make the electron density almost constant. Such a region behaves like a hole in the lattice, and would give iso-diffusion contours having spherical symmetry.

In conclusion, we wish to thank the Director of the Mathematical Laboratory, Dr M. V. Wilkes, for permission to use the EDSAC. One of us (G. K.) thanks the Nuffield Foundation for the award of an Indian Travelling Fellowship in Natural Sciences.

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The Crystal Structure of Cycloserine Hydrochloride*

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The hydrochloride of cycloserine, 4-amino-3-isoxazolidone, is orthorhombic, $P2_12_12_1$, with four molecules in a unit cell of dimensions $a = 9{\cdot}72$, $b = 10{\cdot}34$, $c = 5{\cdot}73$ Å. A successful X-ray analysis was accomplished by means of ($hk0$) and ($h0l$) projections, and three-dimensional syntheses were used to refine atomic coordinates. Bond distances are normal, with the five-membered ring appearing nearly planar. Each chlorine atom forms three weak hydrogen bonds with amino nitrogens, and the ring nitrogen and oxygen have a close approach to the slightly enolized ketone oxygen.

Introduction

Cycloserine is a new broad-spectrum antibiotic produced by *Streptomyces orchidaceus*. The compound was discovered by R. Harned and E. Kropp, and isolated and crystallized by a research team of the Commercial Solvents Corporation (Welch, Putnam & Randall, 1955; Harned, Hidy & Kropp la Baw, 1955). Subsequent chemical and clinical studies on the material have been carried out jointly by groups at the Commercial Solvents Corporation and Eli Lilly and Company. (The name for the compound used by the latter group is Seromycin.)

Cycloserine is an unusual antibiotic, dissimilar in chemical structure with any previously identified, and the culture which produces it is different from any previously known. It is active against both gram-positive and gram-negative bacteria, and seems to differ in its mode of action from other antibiotics presently known. Contrary to *in vitro* results, it has a high degree of *in vivo* efficacy, and is equally effective by oral and subcutaneous routes (Cuckler, Frost, McClelland & Solotorovsky, 1955).

The investigation of the crystal structure of this compound was suggested by Dr Jerome Martin of the Commercial Solvents Corporation, and crystalline material was furnished by him and by Dr Harry Rose of Eli Lilly and Company.

The X-ray analysis completely confirms the chemical elucidation of the antibiotic structure, and provides accurate measurements of bond lengths and angles.

Experimental

Weissenberg photographs of cycloserine reveal the space group as $P2_12_12_1$, with cell constants

$$a = 9{\cdot}72,\ b = 10{\cdot}34,\ c = 5{\cdot}73\ \text{Å}\,.$$

The crystal density, measured by the displacement method using cyclohexane, is 1·598 g.cm.$^{-3}$, from which calculation shows four molecules per unit cell.

X-ray intensity data for the ($hk0$) projection, the ($h0l$) projection, and five layers along the c axis, were obtained from Weissenberg moving-film photographs taken with Cu $K\alpha$ radiation; 616 symmetry-unrelated reflections were recorded with the multiple-film technique, and intensities were estimated visually by comparison with a scale made from a reflection from the same crystal. The c-axis layers were scaled to each other using ($h0l$) data. Lorentz and polariza-

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* Taken from work submitted by J. W. Turley in partial fulfillment of requirements for the degree of Ph.D.