gradients, *etc.*) and the interaction between them will produce an increment in the thickness of the front. The evaporation front then becomes visible. Note for example that in Fig. 6 the thickness is 0.3 micron and in Fig. 2 it is up to 1 micron.

In vacuum, the process is carried out in a different way. The low pressure  $(10^{-5} \text{ torr})$  does not allow the formation of the melt layer on the surface and, thus, the material goes directly from the solid to the vapor phase. The net result is that circular pits are not developed.

It is a pleasure to record the helpful and stimulating discussions with Professor N. Cabrera and H. Riveros, as well as the help of A. Valladares and J. A. Careaga in the preparation of the manuscript.

#### References

- FERNÁNDEZ, A. & MUÑOZ, E. (1962). Rev. Mex. Fís. XI, 255.
- GILMAN, J. J., JOHNSTON, W. G. & SEARS, G. W. (1958). J. Appl. Phys. 29, 747.
- GRINBERG, A. (1963). Phys. Stat. Sol. 3, 1369.
- MENDELSON, S. (1961). J. Appl. Phys. 32, 1579.
- PATEL, A. R., BAHL, O. P. VAGH, A. S. (1965). Acta Cryst. 19, 1025.
- WAINER, L. S. DE & ABELEDO, M. J. DE (1965). Brit. J. Appl. Phys. 16, 1764.

Acta Cryst. (1968). A24, 688

# Diffuse Diffraction Phenomena from Neutron-Irradiated Graphite Single Crystals

# BY W.T. EELES<sup>†</sup>

Berkeley Nuclear Laboratories, Berkeley, Gloucestershire, England

### (Received 4 April 1968)

Single crystals of neutron irradiated graphite have been found to show diffuse diffracted intensity in the form of rel-rods through the hki0 reflexions and parallel to c\*. Off-axis broadening of the 000*l* reflexions has also been observed.

Powder diffraction studies of irradiated graphite (Bacon & Warren, 1956; Eeles, 1962) have shown that radiation damage in graphite results in peak shifts, line broadening and changes in small-angle scattering. These observations are consistent with measurements of other physical properties such as bulk growth and stored energy, and lead to a picture of damage in graphite irradiated at low temperatures (<300 °C) in which displaced carbon atoms are distributed at random, singly or in small clusters, between the layer planes. The layer structure is thus distorted primarily in the c direction. The author has taken oscillation photographs of small single crystals of Ticonderoga graphite irradiated with neutrons to a dose of  $2 \times 10^{20}$ n.v.t. (Ni) at a temperature of 200 °C.

Figs. 1, 2 and 3 are oscillation photographs of the irradiated crystals, while Figs. 4 and 5 are oscillation photographs of unirradiated crystals of comparable size, taken under identical conditions to those of Figs. 1 and 2. Comparison of Figs. 1 and 4 shows that neutron irradiation has caused extensive asymmetric axial broadening of the 000l reflexions. These reflexions are also diffuse in directions at right angles to  $c^*$ . That may simply be because of macroscopic mis-orientated regions which often occur in Ticonderoga graphite, or

it may be spreading of the reflexions in reciprocal space. The 0002 reflexion is in fact so diffuse that it appears in photographs taken with c as axis of oscillation (Fig. 2).

Fig. 2 shows another striking feature when compared with Fig. 5, viz. continuous rods of diffuse intensity on the row lines passing through each hki0 reflexion. The rods are shown in cross-section in Fig. 3, which is a stationary crystal photograph taken with **c** parallel to the incident X-ray beam. **c**-Axis strain could cause extension of the *hkil* reflexions but it cannot give rise to the diffuse intensity around the *hki0* reflexions since the reciprocal lattice vectors for these reflexions are at right angles to the strain. **a**-Axis strains would broaden these reflexions along the *hki0* reciprocal lattice vectors, not at right angles to them as in the present case.

Factors which are known to extend the hki0 reflexions parallel to  $c^*$  are (Wilson, 1949) stacking faults in which adjacent layers are displaced relative to one another by  $\frac{1}{3}$ [1120], and twist boundaries (turbostratic disorder) in which adjacent layers are rotated with respect to one another about the c axis by random amounts. The first kind of fault would not affect 1120 reflexions at all, while the second kind would give diffuse intensity as cylinders with  $c^*$  as axis, rather than rods (*cf.* Fig. 3). A third type of fault, translation of layers by random amounts in the [1120] direction, is consistent with rods passing through all hki0 reflexions

<sup>†</sup> Present address: Electricity Council Research Centre, Capenhurst, Chester, England.

# ACTA CRYSTALLOGRAPHICA, Vol. A 24, 1968-Eeles



Fig. 1. Neutron-irradiated graphite,  $30^{\circ}$  oscillation photograph, 10T0 as axis of oscillation. 000/ reflexions on the zero layer (Mo K $\alpha$  X-rays).



Fig. 2. Neutron-irradiated graphite,  $60^{\circ}$  oscillation photograph, c as axis of oscillation. Zero layer reflexions: innermost 10T0, outermost 11Z0. (Cu K $\alpha$  X-rays).



Fig. 3. Neutron-irradiated graphite, stationary crystal photograph with c parallel to the incident X-ray beam (Mo $K\alpha$  X-rays).



Fig. 4. Unirradiated graphite. Orientation and exposure conditions as for Fig. 1.



Fig. 5. Unirradiated graphite. Orientation and exposure conditions as for Fig. 2.

but must be rejected since there seems to be no mechanism by which neutron bombardment could produce such faults.

However, there appear to be three plausible ways in which neutron bombardment could result in the observed rod-like extension of the hki0 reflexions:

(a) The clustering of vacant lattice sites within a layer to form disc-like voids, which if uncollapsed would be one interlayer spacing thick in the c direction.

(b) The creation of voids between the layers as a result of the insertion of interstitial atoms either singly or in very small clusters. The relatively weak bonding between layers in graphite, in conjunction with the very strong bonding in a layer, makes it reasonable to suppose that when adjacent layers are forced apart they do not relax to the normal inter-layer spacing for some considerable distance from the interstitial atom.

(c) The subdivision of the crystal into small domains, in themselves undistorted, but having no coherent phase relationship with one another because they are separated by regions distorted by the insertion of interstitial atoms. To account for the diffuse rods the domains would have to be much thinner in the cdirection than at right angles to it.

The first hypothesis must be rejected because it has been established (Simmons, 1965) that neutron bombardment creates vacancies singly and at random through the lattice, and that they are immobile until temperatures greatly in excess of 300 °C are reached. Interstitial atoms are sufficiently mobile below 300 °C to be able to diffuse within any one interlayer space, and they may thus minimize the strain in the lattice around them by forming small, less mobile clusters.

Hypotheses (b) and (c) are not mutually exclusive and both should lead to diffuse rod-like scattering

around all reflexions, including 0000 and the 000l reflexions. Inhomogenous c-axis strain should, however, result in the reflexions with  $l \neq 0$  being broadened to different extents. Diffuse scattering along  $c^*$  in the vicinity of 0000 has previously been observed with polycrystalline irradiated graphite but in the present experiments cannot be separated from the white radiation streak of the 0002 reflexion with any certainty. Hypothesis (c) is consistent with the suggestion of Bacon & Warren (1956) that the very anisotropic strain around an interstitial atom or cluster is such that a pair of interstitials in close proximity within the same interlayer space tend to amalgamate, while interstitial atoms in close proximity but in different interlayer spaces repel one another. Such migration as a result of mutual interaction could leave small regions of undistorted lattice between regions containing displaced atoms.

To distinguish between the relative contributions of strain and the two hypotheses advanced here requires a more quantitative theoretical analysis and more precise experimental measurements, which are not possible at the present time. The author is indebted to Professor A.J.C. Wilson and Dr G.K. Williamson for very helpful discussions and to the Central Electricity Generating Board for permission to publish the photographs.

#### References

BACON, G. & WARREN, B. E. (1956). Acta Cryst. 9, 1029. EELES, W. T. (1962). Properties of Reactor Materials and

the Effects of Radiation Damage. Edited by D. J. Littler. London: Butterworths.

SIMMONS, J. W. (1965). Radiation Damage in Graphite. Oxford: Pergamon Press.

WILSON, A. J. C. (1949). X-ray Optics. London: Methuen.

### Acta Cryst. (1968). A 24, 689

# The Relationship between X-ray Scattering and the Mössbauer Effect: a Fourier Approach

# By J.A. Elliott

Physics Department, University of Manchester Institute of Science and Technology, Manchester 1, England

### (Received 9 January 1968 and in revised form 3 May 1968)

The well-established relationship between X-ray scattering and the Mössbauer effect is demonstrated by deriving the main features of both phenomena as the Fourier transform of a time-dependent Patterson function.

The close correspondence between the Mössbauer effect and the scattering of X-rays has been pointed out by many authors. Frauenfelder (1962) discusses the Debye-Waller temperature factor as it occurs in both X-ray and Mössbauer theory. Tzara (1961) gives a more detailed treatment, and Lipkin (1961) points out the general occurrence of the Debye-Waller factor in any interaction involving an impulsive momentum transfer to a crystal lattice.

It is the purpose of this note to discuss this close relationship using the fundamental tools of X-ray crystallography, the Fourier transform, the convolution theorem, and the Patterson function. The argument is semi-qualitative, and draws attention to the physical principles involved, from the viewpoint of X-ray crystallography.

The essence of the Mössbauer effect is the recoilless emission of  $\gamma$ -rays from certain atomic species in suitable environments, such as crystalline solids: the radiation exhibits no energy shift due to recoil of the emitting nucleus, and also exhibits no Doppler broadening. The energy width of the radiation is, therefore,