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 c direction 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.

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

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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 γ-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,

that of the nuclear transition only. In the case of [57]Fe, the most commonly used Mössbauer isotope, this gives rise to 14·4 keV γ-rays having a width of only 10^{-8}eV, a resolution of 10^{-12}. (See, for example, Wertheim, 1964.)

In each process there is an impulsive momentum transfer to the lattice. The impulse is absorbed not by the excitation of lattice modes, but by the phonon-free recoil of the whole crystal. In the Mössbauer case, the nucleus emits a γ-ray of energy hcS_o ($S_o=2\pi/\lambda$) and a corresponding momentum hS_o is imparted to the lattice. In the X-ray case, the change of momentum $h(S-S_o)\equiv hs$ of the X-ray is transmitted to the lattice: the effect on the lattice is the same. The relationship between the two processes is well illustrated by taking a Fourier transform approach. For simplicity, we shall consider a simple monatomic lattice.

It is well known in X-ray studies that the intensity distribution in reciprocal space of the scattered radiation is given by the Fourier transform of the spacial distribution function $\mathscr{P}(\mathbf{x})$, the Patterson function. This relationship can be extended into the fourth dimension, in that the energy distribution of the scattered radiation is the transform of the time dependence of the distribution function. More specifically (following the notation of Guinier, 1963), the intensity distribution as a function of momentum hs and energy hv is given by the space-time transform of the correlation function $\mathscr{P}(\mathbf{x}, t)$:

$$I_N(\mathbf{s}, v)=\int \mathscr{P}(\mathbf{x}, t) \exp [2\pi i(vt-\mathbf{s}.\mathbf{x})]dv_xdt . \quad (1)$$

hv is the energy change of the photon on scattering. $\mathscr{P}(\mathbf{x}, t)$, the time-dependent pair correlation function (introduced by van Hove, 1954), measures the probability that, given an atom at $x=0$ at $t=0$, there will be an atom (not necessarily the same one) at x at time t. In this we have assumed, for convenience, point atoms, i.e. atomic scattering factors equal to unity. For finite atoms, the atomic scattering factor appears simply as a factor $|f(\mathbf{s})|^2$ which carries through.

Defining $\mathscr{Q}(\mathbf{s}, t)$ as an intermediate correlation function, we have the following transform relationships:

$$\mathscr{P}(\mathbf{x}, t) \leftrightarrow \mathscr{Q}(\mathbf{s}, t) \leftrightarrow I_N(\mathbf{s}, v) . \quad (2)$$

In conventional X-ray practice, energy discrimination of the required precision is not possible owing to the large width of the characteristic lines; what is measured, therefore, is $\int I_N(\mathbf{s}, v)dv$. Clearly, on transformation this yields $\mathscr{P}(\mathbf{x}, 0)\equiv\mathscr{P}(\mathbf{x})$ the instantaneous distribution function, or Patterson function. The significance of the relationships (2) will be considered below.

Similar Fourier relationships occur in the Mössbauer case (Elliott, Hall & Bunbury, 1966) with the important difference that, being an emission process, and therefore essentially incoherent, no information concerning interatomic correlation can be obtained. Thus, only the self-correlation function $\mathscr{P}_s(\mathbf{x}, t)$ is involved, describing the motion of a single (average)
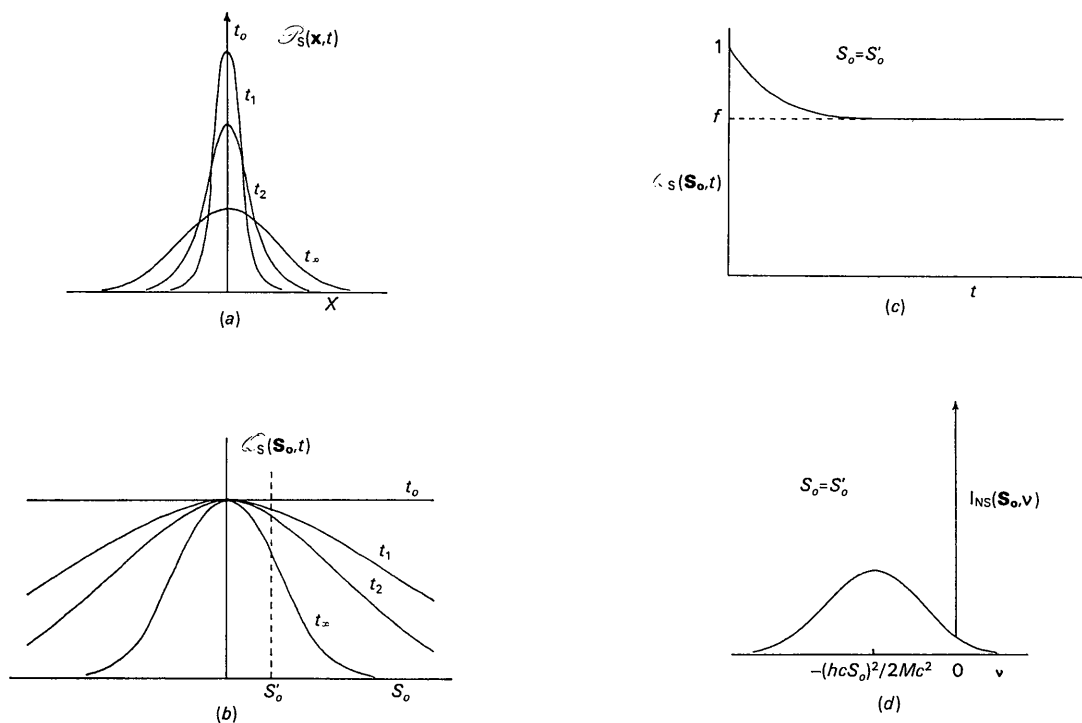

Fig. 1. (a) The distribution function $\mathscr{P}_s(x, t)$ as a function of x, showing its spreading with time. (b) The space Fourier transform of $\mathscr{P}_s(x, t)$ corresponding to the curves in Fig. 1(a). (c) $\mathscr{Q}_s(S_o, t)$ plotted as a function of t for a given value of S_o. (d) The time Fourier transform of $\mathscr{Q}_s(S_o, t)$ for the value $S_o=S_o'$. $I_{NS}(S_o, v)$ is thus the space–time transform of $\mathscr{P}_s(x, t)$.

atom. $\mathscr{P}_s(\mathbf{x}, t)$ is the probability that, given an atom at $x=0$ at $t=0$, the same atom will be at x at time t. The emission spectrum in the Mössbauer case is therefore given by

$$I_{Ns}(\mathbf{S}_o, v) = \int \mathscr{P}_s(\mathbf{x}, t) \exp [2\pi i(vt - \mathbf{S}_o, \mathbf{x})]dv_x dt \quad (3)$$

(ignoring nuclear effects), and corresponding to the relationships (2) we have

$$\mathscr{P}_s(\mathbf{x}, t) \leftrightarrow \mathscr{Q}_s(\mathbf{S}_o, t) \leftrightarrow I_{Ns}(\mathbf{S}_o, v) . \quad (4)$$

(note that since the energy changes involved are very small, $\sim kT$, $\mathbf{S}_o$ and v can be regarded as independent variables).

In a typical solid, $\mathscr{P}_s(\mathbf{x}, t)$ has the behaviour shown in Fig. 1(a). By definition, at $t=0$ it is a δ-function in x. At later times, it broadens (corresponding to atomic thermal motion) until at $t \sim 10^{-12}$ seconds it has settled down to a Gaussian shape of constant width, describing the probability distribution for an atom under thermal motion. Fig. 1(b) and (c) give the corresponding function $\mathscr{Q}_s(\mathbf{S}_o, t)$ as a function of $\mathbf{S}_o$ and t respectively, derived qualitatively from simple Fourier transform properties. $\mathscr{Q}_s(\mathbf{S}_o, t)$ transforms to give $I_{Ns}(\mathbf{S}_o, v)$, the general behaviour of which is shown in Fig. 1(d). Its main features are a very narrow peak, the Mössbauer radiation, which a more detailed analysis shows to occur at $v=0$ (*i.e.* it corresponds to recoilless emission), and a broad peak occurring at the shifted position, $hv = (hcS_o)^2/2Mc^2$ corresponding to emission with recoil and Doppler broadening (M is the nuclear mass). The origins of the two peaks are clearly seen in this treatment: the Mössbauer peak occurs because the atom is confined to oscillate about a fixed lattice site, and the broad inelastic peak occurs as a result of thermal motion.

This treatment can be made semi-quantitative. Since $\mathscr{P}_s(\mathbf{x}, \infty)$ is a Gaussian, we can write

$$\mathscr{P}_s(\mathbf{x}, \infty) = (2\pi a^2)^{-3/2} \exp (-x^2/2a^2) \quad (5)$$

and therefore

$$\mathscr{Q}_s(\mathbf{S}_o, \infty) = \exp (-\tfrac{1}{2}S_o^2 a^2) . \quad (6)$$

An examination of Fig. 1 will show that $\mathscr{Q}_s(\mathbf{S}_o, \infty)$ is the area of the Mössbauer peak, and is thus the recoilless fraction $f = e^{-2M}$. This is identical with the Debye–Waller temperature factor of X-ray work. We shall now show that in the X-ray case, the sharp peak corresponds to elastic scattering, and the broad peak to thermal diffuse scattering.

For the X-ray case, we consider not $\mathscr{P}_s(\mathbf{x}, t)$ but $\mathscr{P}(\mathbf{x}, t)$, which includes pair correlations and therefore coherence effects. The form of $\mathscr{P}(\mathbf{x}, t)$ can easily be derived quantitatively for a crystalline solid. It must have a δ-function at the origin at $t=0$, spreading like $\mathscr{P}_s(\mathbf{x}, t)$ at later times. The probability distribution around other lattice sites is constant in time, and equal to $\mathscr{P}_s(\mathbf{x}, \infty)$. The distribution of sites is given by $\mathscr{P}(\mathbf{x})$. Hence

$$\mathscr{P}(\mathbf{x}, t) = \mathscr{P}(\mathbf{x}) * \mathscr{P}_s(\mathbf{x}, \infty) + \mathscr{P}_s(\mathbf{x}, t) - \mathscr{P}_s(\mathbf{x}, \infty) , \quad (7)$$

where $*$ signifies a convolution (faltung).

Thus

$$\mathscr{Q}(\mathbf{s}, t) = I_N(\mathbf{s})\mathscr{Q}_s(\mathbf{s}, \infty) + \mathscr{Q}_s(\mathbf{s}, t) - \mathscr{Q}_s(\mathbf{s}, \infty) \quad (8)$$

and

$$I_N(\mathbf{s}, v) = [I_N(\mathbf{s})e^{-2M}\delta(v)] + [I_{Ns}(\mathbf{s}, v) - e^{-2M}\delta(v)] \quad (9)$$

giving the distribution of the scattered radiation in both reciprocal space and energy.

The first term of equation (9) is a δ-function in energy, and describes elastic scattering, occurring at Bragg angles only, with an intensity determined by e^{-2M}, the temperature factor. It corresponds to the recoilless Mössbauer peak, and arises for the same reason *i.e.* the confinement of the atom. The second term is just the broad peak of Fig. 1(d), and gives inelastic scattering at all angles – the thermal diffuse scattering (*cf.* James, 1947).

It is thus demonstrated that elastic X-ray scattering and the Mössbauer effect have a close correspondence, and occur under the same conditions of temperature, wavelength *etc.* since their intensities are governed by the same factor e^{-2M}. The problems associated with the observation of the effects are, of course, quite different, and considerably more severe in the Mössbauer case.

It is worth noting that although energy analysis of the required precision cannot be achieved in conventional methods, analysis of the elastic peak can be achieved if a Mössbauer source is used instead of an X-ray generator. The energy analysis is then performed by using conventional Mössbauer techniques (Wertheim, 1964). O'Connor & Butt (1963) have performed experiments in this way, and succeeded in separating the elastic peak (the first term of equation (9)) from the inelastic background. Elliott *et al.* (1966), scattering from a viscous liquid, showed the effect on the elastic peak of diffusive atomic motion in the scatterer. The peak becomes broader in energy (this is essentially the Doppler effect) in a manner depending on the diffusive mechanism. However, the scattered intensities obtainable in such experiments are extremely low compared with those from conventional X-ray tubes.

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