A Method for the Interpretation of the Warren–Averbach Mean-Squared Strains and Its Application to Recovery in Aluminium

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Abstract
A method for the interpretation of Warren–Averbach mean-squared strain curves is presented. If a polynomial is fitted to such a mean-squared strain curve, then the coefficients of this polynomial can be expressed simply in the mean-squared local strain in the direction perpendicular to the diffracting planes and the mean-squared derivatives of this strain. The method is applied to a series of X-ray diffraction line broadening measurements of isochronally recovery-annealed aluminium. The changes in the average squared derivatives observed after different anneals are explained as changes in the dislocation arrangements in the specimen. The parameters obtained by the method are found to be sensitive to small changes in the strain fields.

1. Introduction
Many important properties of materials depend critically on the density, nature and exact arrangement of dislocations in the crystals. At present there are a number of ways to study dislocations experimentally, even though all methods contain some limitations or difficulties in carrying out the experiments or in the interpretation of the results. One of the methods is to measure X-ray diffraction line broadening. Although the line broadening is not caused by the dislocations themselves but by the accompanying strain fields, it should in principle be possible to make deductions on the density and the general arrangement of the dislocations from X-ray diffraction line profiles.

A generally used method to interpret X-ray diffraction line broadening is that given by Warren & Averbach (cf. Warren, 1969), which yields information on the non-uniform strains in the form of mean-squared strains as a function of correlation length. For the interpretation of these mean-squared strains no totally satisfactory procedure is known at present, beyond an estimation of dislocation density.

In the present contribution first an approach is presented to interpret the mean-squared strain curves as obtained from the Warren–Averbach analysis. Then it is applied to a series of X-ray diffraction line profile measurements on cold-worked and subsequently recovery-annealed aluminium, i.e. on a material containing different dislocation arrangements at a practically constant average dislocation density.

2. Theory
By applying the Warren–Averbach analysis the structural broadening of X-ray diffraction peaks from cold-worked metals can be divided into two parts: one part is dependent on the order of the reflection, while the other is not. Accordingly, the real part of the Fourier transform, A, of the structural intensity distribution can be written as

\[ A = A_S A_P, \]

where \( A_S \) expresses the so-called size broadening which is independent of the order, while the order-dependent factor \( A_P \) represents the strain broadening.

The strain broadening, \( A_P \), depends on the non-uniform strains present in the crystals in the following manner:

\[ A_P(L) = \left< \cos \frac{2\pi L \delta(L)}{d} \right> \geq 1 - 2\pi^2 \left( \frac{L}{d} \right)^2 \left< \delta(L)^2 \right>, \]

where \( L \) is a distance in the crystal perpendicular to the reflecting lattice planes and \( d \) is the spacing of the reflecting planes, which depends on the order of the reflection. The function \( \delta(L) \) gives the strain perpendicular to the reflecting planes averaged over a distance of length \( L \) [see (3)]. The brackets in (2) denote averaging with respect to all ranges of length \( L \) to be found in all the coherently diffracting columns contained in the crystals. The mean-squared strain for different distances \( L \) occurring in (2) can be written as

\[ \left< \delta(L)^2 \right> = \left< \left[ \frac{1}{L} \int_{-L/2}^{L/2} E(\zeta) d\zeta \right]^2 \right>, \]

where \( \zeta = z - z_o \), where \( z \) is a coordinate in the diffraction-
ing crystal perpendicular to the reflecting planes and $z_0$ is the middle point of a particular range of length $L$ in a column. The averaging, indicated by the brackets, $\langle \cdot \rangle$, in (3) is thus with respect to all possible values of $z_0$ contained in all the diffracting columns. The normal strain as a function of $z$ is denoted by $E(z)$ to distinguish it from the function $e(z)$, which gives the local strain as a function of the actual coordinate, $z$. For every range of length $L$ there exists a function $E(\zeta)$ defined in the interval $-L/2 \leq \zeta \leq L/2$ so that $E(\zeta) = e(z - z_0)$.

Since $E^{(n)}(0) = e^{(n)}(z_0)$, where $E^{(n)}$ and $e^{(n)}$ are $n$th derivatives, the function $E(\zeta)$ can be represented by the following Taylor series.

$$E(\zeta) = e(z_0) + \sum_{n=1}^{\infty} \frac{e^{(n)}(z_0)}{n!} \zeta^n.$$  \hspace{1cm} (4)

When this series is integrated from $-L/2$ to $L/2$ and divided by $L$, the odd powers of $L$ vanish, and we obtain

$$e(L) = e(z_0) + \sum_{n=1}^{\infty} \frac{e^{(2n)}(z_0)}{(2n+1)!} 2^{2n} L^{2n}.$$  \hspace{1cm} (5)

After this series is squared and the average is taken over all possible values of $z_0$, the mean-squared strain can be expressed in the following form:

$$\langle e(L)^2 \rangle = C_0 + C_2 L^2 + C_4 L^4 + C_6 L^6 + C_8 L^8 + \ldots$$

$$= \sum_{n=0}^{\infty} C_{2n} L^{2n},$$  \hspace{1cm} (6)

where the first five coefficients have the expressions

$$C_0 = \langle e(z)^2 \rangle$$

$$C_2 = \frac{\langle e(z) e''(z) \rangle}{12}$$

$$C_4 = \frac{\langle e(z) e^{(iv)}(z) \rangle + \langle e''(z)^2 \rangle}{960} + \frac{\langle e''(z)^2 \rangle}{576}$$

$$C_6 = \frac{\langle e(z) e^{(vi)}(z) \rangle + \langle e(z) e^{(iv)}(z) \rangle}{161280} + \frac{\langle e'(z)^2 \rangle}{23040}$$

$$C_8 = \frac{\langle e(z) e^{(vii)}(z) \rangle + \langle e(z) e^{(v)}(z) \rangle}{46448640} + \frac{\langle e'(z)^2 \rangle}{3870720} + \frac{\langle e^{(iv)}(z)^2 \rangle}{3686400}.$$  \hspace{1cm} (7)

The averaging above was carried out with respect to $z_0$, which means that the columns are not included completely in it, but a range of length $L/2$ at both ends is excluded. Since $L$ in the Warren–Averbach analysis is considerably smaller than the length of a column, this has a negligible effect on the values of the averages, if the general behaviour of the function $e(z)$ at the ends of the columns is the same as elsewhere.

The physical factors that determine the exact column structure of the diffracting crystals, i.e. the length and relative positions of the coherently diffracting columns, are not known very well. The assumption that the behaviour of the function $e(z)$ does not change at the column ends means that there is no correlation between the strain and the factors controlling the column structure. In other words, the division of the crystals into columns is random with respect to the strain. Although it may not be strictly true in all cases, it is certainly a physically realistic assumption. The experimental results of the present work support it.

If the assumption of the randomness of the strains at the column ends is accepted, then the averages of the normal strain, $e(z)$, and of all its derivatives, $e^{(n)}(z)$, at the column ends vanish.

If this is taken into account and repeated partial integration is applied to the terms in (7), it turns out that the coefficient, $C_{2n}$, can be expressed as

$$C_{2n} = \frac{2(-1)^n}{(2n+2)!} \langle e^{(2n)}(z)^2 \rangle.$$  \hspace{1cm} (8)

Accordingly, the first five terms of the expression for the mean-squared strain function will be

$$\langle e(L)^2 \rangle = \langle e(z)^2 \rangle - \frac{1}{12} \langle e'(z)^2 \rangle L^2 + \frac{1}{360} \langle e''(z)^2 \rangle L^4$$

$$- \frac{1}{20160} \langle e'''(z)^2 \rangle L^6$$

$$+ \frac{1}{1814400} \langle e^{(iv)}(z)^2 \rangle L^8 - \ldots.$$  \hspace{1cm} (9)

Since the $\langle e(L)^2 \rangle$ curve is obtained through the Warren–Averbach analysis from the measurements, it is possible, by fitting (9) with a sufficient number of terms to it, to determine in addition to the actual $\langle e(z)^2 \rangle$ the averages of all squared derivatives of the strain. These quantities are directly connected to the strain fields that are present in the diffracting crystals, and it can be expected that for different types of dislocation configurations their relative values will differ.

Eastabrook & Wilson (1951) have given a series expression for the displacements in a column, which is comparable to our (4). Their series is developed with respect to the end point of each range of length $L$ and not with respect to the middle point, $z_0$, as here. However, if their approach is developed further in the same way with the same assumption as above, a result identical to (9) is obtained.

In (2) the higher-order terms are omitted. In principle, it should be possible to develop the higher powers of $e(L)$, in the same way as above, and to use them in a more detailed analysis.

### 3. Experimental

Commercial cold-rolled 2 mm thick aluminium sheet was used for the experiments. The purity of the metal
was 99.5%; most important impurities being Fe and Si. Because deformed aluminium has a negligible density of stacking faults and twins the observed X-ray diffraction line broadening will be caused almost entirely by the dislocations present in the material. Also, the relatively large penetration depth in aluminium for the X-rays applied is significant, since thereby the line broadening better reflects the dislocation structure in the bulk of the material.

Two specimens, in the form of circular disks with a diameter of 50 mm, were cut from the same sheet. The face of the specimens to be used in the diffraction measurements was at first ground lightly, to remove dirt and surface roughness, and then approximately 0.1 mm of surface material was removed by etching for 0.5 h in a solution of 1.5% HCl, 2.5% HNO₃ and 0.5% HF in water. One specimen was annealed, in air, for 0.5 h at successively rising temperatures between 360 and 574 K,* with approximate intervals of 20 K. The specimen was placed inside the furnace on a large aluminium block, which had been stabilized at the required temperature beforehand. It took approximately 10 min for the specimen to reach the desired temperature. The annealing time, 0.5 h, was counted from the moment the specimen was placed into the furnace to the moment it was taken out and allowed to cool in air. The annealing temperatures given here are the maximum temperatures the specimen reached (±3 K). The other specimen was recrystallized in the same furnace for 0.5 h at 673 K for use as a reference. In the calculations 2 sinθ/λ was chosen at the centroids of the line profiles; therefore the structural broadening only were then subjected to a Warren–Averbach analysis, thereby applying the procedure described by Delhez & Mittemeijer (1976), i.e. making use of a linear relationship between the Fourier coefficients themselves and the square of the reflection instead of a linear relationship

*Annealing at higher temperatures resulted in partial recrystallization as was detected by marked changes in the relative diffraction peak intensities; between 360 and 574 K very little change in intensities was observed.

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The instrumental broadening was removed from the net measured profiles by the Stokes deconvolution procedure (Stokes, 1948), using the recrystallized specimen as a reference. In the calculations 2 sinθ/λ was used as an argument for the intensity distributions, θ being the diffraction angle and λ the wavelength of the X-rays. In the Fourier series evaluation origins were chosen at the centroids of the line profiles; therefore the determination of the Warren–Averbach analysis refer to the average lattice spacing of the specimens (de Keijser & Mittemeijer, 1980). The Fourier coefficients of the 200 and 400 line profiles reflecting the structural broadening only were then subjected to a Warren–Averbach analysis, thereby applying the procedure described by Delhez & Mittemeijer (1976), i.e. making use of a linear relationship between the Fourier coefficients themselves and the square of the reflection instead of a linear relationship

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**Fig. 1.** Quadrants of the 111 pole figures from the specimen in its original cold-worked state and from the recrystallized reference specimen.
between the logarithm of the Fourier coefficients and the square of the order. For comparison the logarithmic relation was applied to a few sets of reflections, and it was found that both procedures give practically the same results. Furthermore, the \( \langle \sigma(L)^2 \rangle \) values obtained (cf. Fig. 3) show that (2) is a sufficiently accurate approximation, even for the largest values of \( L \) used in the present analysis. A power series of \( L^2 \) of the type (6) was fitted to the \( \langle \sigma(L)^2 \rangle \) curve obtained. The first twelve terms of the series were taken into account and the fit was extended over the first 24 values of \( L \), i.e. from 22 to 536 Å. From the coefficients \( C_{2n} \), values of \( \langle e(L)^2 \rangle \) and of the mean-squared strain derivatives \( \langle e''(z)^2 \rangle \) were determined according to (8).

4. Results

Figs. 2 and 3 give the size Fourier coefficients and the \( \langle \sigma(L)^2 \rangle \) curves obtained from the measurements for a few annealing temperatures. Some systematic changes can be observed in both sets of curves. The size coefficient curves seem to become gradually more concave at higher temperatures (cf. Discussion) while in the \( \langle \sigma(L)^2 \rangle \) curves the values for larger \( L \) clearly diminish with the increase of the annealing temperature. The values of \( \langle \sigma(L)^2 \rangle \) for small \( L \) do not seem to change appreciably.

The changes in the \( \langle \sigma(L)^2 \rangle \) curves can be further analysed with the aid of the average squared derivatives obtained from the fitting procedure described in § 3. It was found, however, more helpful not to study the absolute values of these averages, but the ratios of two successive ones, such as \( \langle e'(z)^2 \rangle / \langle e(z)^2 \rangle \), \( \langle e''(z)^2 \rangle / \langle e'(z)^2 \rangle \), \( \langle e'''(z)^2 \rangle / \langle e''(z)^2 \rangle \), etc. There are two advantages in using these quantities, firstly, the ratios show the change in a particular average with respect to a constant level of the average of one step lower derivative and, secondly, the effects of statistical errors in the \( \langle \sigma(L)^2 \rangle \) curves are diminished. The mean-squared local strain \( \langle e(l)^2 \rangle \) obtained from the fitting and the ratios for the first three average squared derivatives versus the annealing temperature are seen in Fig. 4. For the examples of the mean-squared strain curves shown in Fig. 3 the numerical results of the fitting are shown in Table 1.

In the fitting of an expression of type (6) to a mean-squared strain curve there is a danger of serious errors, if the fitting is not done carefully enough. In the present work in all cases 12 terms in (6) were taken into account in a direct least-squares fit. An alternative is to use some intermediate functions, such as \( \exp(-nL^2) \). The number of terms necessary for a good fit depends on the maximum value of \( L \) to be included and on the shape of the \( \langle \sigma(L)^2 \rangle \) curve. The actual number of terms in each particular case is best ascertained by comparing the fitted values to the measured ones. It was found, however, that at least ten terms must be included in any practical case. It seems also to be good practice to have the number of terms about one half of the number of experimental points.

Fig. 2. The size Fourier coefficients for three examples. The curve for 477 K is almost identical to the one for 400 K.

Fig. 3. The mean-squared strain values as obtained from the measurements (dots) and the curves fitted to them for the four examples.
Table 1. The coefficients $C_{2n}$ for the four examples seen in Fig. 3 as well as the corresponding average squared local strain derivatives and their ratios

Also the halfwidths, $W$, of the measured 200 and 400 $K_2$ peaks are shown. For the reference peaks $W = 0.091$ and 0.136 for 200 and 400, respectively.

<table>
<thead>
<tr>
<th>Annealed at</th>
<th>Not annealed</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>400 K</td>
</tr>
<tr>
<td>$W_{200}$ (20)</td>
<td>0.113</td>
</tr>
<tr>
<td>$W_{400}$ (20)</td>
<td>0.238</td>
</tr>
<tr>
<td>$C_0 \times 10^3 \ A$</td>
<td>4,965</td>
</tr>
<tr>
<td>$C_2 \times 10^{10} \ A^2$</td>
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</tr>
<tr>
<td>$C_4 \times 10^{20} \ A^4$</td>
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</tr>
<tr>
<td>$C_6 \times 10^{20} \ A^8$</td>
<td>0.8981</td>
</tr>
<tr>
<td>$C_{10} \times 10^{24} \ A^{10}$</td>
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</tr>
<tr>
<td>$C_{12} \times 10^{35} \ A^{12}$</td>
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</tr>
<tr>
<td>$C_{14} \times 10^{40} \ A^{14}$</td>
<td>-14.13</td>
</tr>
<tr>
<td>$C_{16} \times 10^{45} \ A^{16}$</td>
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</tr>
<tr>
<td>$C_{18} \times 10^{51} \ A^{18}$</td>
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</tr>
<tr>
<td>$C_{20} \times 10^{55} \ A^{20}$</td>
<td>11.61</td>
</tr>
<tr>
<td>$C_{22} \times 10^{63} \ A^{22}$</td>
<td>-17.61</td>
</tr>
</tbody>
</table>

The fitted curves in Fig. 3 follow very accurately the experimental points at low values of $L$, but at the high end some oscillations can be seen. These are caused by neglecting the higher-order terms in the series (6), however, they are unimportant, if only the values of $\langle e(z)^2 \rangle$ and the lower-order mean-squared derivatives are desired.

In three cases, namely with the unannealed specimen and at temperatures 439 and 538 K, the fit gave negative values for some of the average squared derivatives. In all other cases the first 12 coefficients $C_{2n}$ in the series (6) turned out to be alternately positive and negative (see Table 1), as would be expected [cf. (8)]. The anomaly was strongest in the measurement after the 439 K anneal (omitted from the figures). After careful examination of conditions in which this measurement was carried out as well as of the diffraction data and its subsequent analysis, no clear explanation for this deviation was discovered. It can only be concluded that the deviations are caused by cumulative errors in the measurement and in the analysis of the data [leading to too low values of $\langle e(L)^2 \rangle$ for small $L$]. The deviations in the two other cases mentioned were smaller and occurred in higher terms, but they obviously originate from the same reasons. It can be seen in Fig. 4 that $\langle e(z)^2 \rangle$ changes very little during this series of anneals, only a small decrease can be observed. If the dislocation density is estimated on the basis of the value of $\langle e(z)^2 \rangle$, according to the method of Williamson & Smallman (1956) we end up with the approximate value of $10^{10} \ cm^{-2}$, which is quite reasonable (cf. Williamson & Smallman, 1956). The curves in Fig. 4 picturing the ratios of the first three average squared derivatives of the strain are very similar. It is clear that there is at first a marked increase in the values of all three quantities, and after about 473 K a decrease. Further, it is intriguing that the ratios $\langle e^{(n)}(z)^2 \rangle/\langle e^{(n-1)}(z)^2 \rangle$ for $n \geq 2$ have approximately the same value (see also Table 1). This is not understood at present. The changes in the averages of the squared strain derivatives are almost certainly connected to changes in the dislocation configurations in the specimen. These changes are quite small, however. In order to obtain supporting information for these results, a number of specimens were made from the same cold-worked material and annealed in the same way, after which they were studied in a transmission electron microscope. No clear differences between specimens annealed at different temperatures could be resolved.
5. Discussion

The fitting of the series expansion (6) to the $\langle d(L)^2 \rangle$ curves obtained from the Warren–Averbach analysis provides a useful method for extracting information about the strain field in the specimen. It differs from previous techniques in that it has a more rigorous physical basis, since the quantities obtained here have a straightforward connection to the strain fields in the diffracting crystals (§ 3).

Of the previous attempts to interpret Warren–Averbach mean-squared strain curves the most recent and perhaps the most successful one is from Adler & Houska (1979). They approximate the $\langle d(L)^2 \rangle$ curve by a function proportional to $1/(L^2r)$, where the exponent $r$ varies from 0.38 to 0.61. The same expression, with $r = 0.5$, was previously used by Rothman & Cohen (1969). The results of our measurements, however, show significant systematic differences from this relation.

The reliability of the results obtained from the described fitting procedure depends greatly on the errors present in the mean-squared strain curves as obtained from the Warren–Averbach analysis. In particular, the errors at the low-$L$ part of the $\langle d(L)^2 \rangle$ curve affect the coefficients $C_n\eta$ and, consequently, the average squared derivatives calculated from them. In connection with that, two types of errors have to be considered: (i) errors due to an incorrect estimation of background, and truncation of the line profile, and (ii) errors due to assumptions in the Warren–Averbach method. Incorrect background levels lead to errors in the Fourier coefficients with small harmonic numbers, thereby causing errors in the normalization of the Fourier coefficients, as needed in the Warren–Averbach analysis. These errors propagate in the Warren–Averbach analysis in such a way that the mean-squared strains for small $L$ are affected and that the size coefficients, $S_n(L)$, may show a negative curvature at small $L$, which is known as the ‘hook effect’. The present experiments were carried out very carefully: errors due to counting statistics were small and the measurements were extended over wide $2\theta$ ranges in order to include the tails of the line profiles and to be able to determine the background reliably. Moreover, the background was very low compared with the maximum peak intensities and, due to the texture (Fig. 1), the intensities of the 200 and 400 line profiles were relatively high compared with neighbouring reflections, which means that overlap of profile tails could not play an important role. The observed negative curvature of $A^2(L)$ (Fig. 2) could not be explained by estimating the maximum possible effect on the basis of the background versus peak intensity ratio (see, for example, Cheary & Grimes, 1972). The part of the broadening determined in the Warren–Averbach analysis that is frequently called the size broadening (which includes broadening due to stacking faults and twins) is in fact the order-independent part of the broadening and there is no need to interpret it fully as size broadening. Other structural defects may contribute to it and may cause the negative curvature. It should also be noted that Wilkens (1979) has reported theoretical simulations of Warren–Averbach analysis, which resulted in size coefficients that show a negative curvature. Therefore, no attempt whatsoever was made to correct for the hook effect in the present investigation and we conclude that, within the limits of the reliability of the Warren–Averbach method (Warren, 1969; Delhez & Mittemeijer, 1976), the experimental errors are not the main cause of the hook effect observed and that the mean-squared strain curves obtained are trustworthy. The changes in the average squared derivatives of the strain as seen in Fig. 4 can be interpreted as indications.
of changes in the dislocation structure of the specimen.

In a quite recent investigation of Hasegawa & Kocks (1979) two types of recovery behaviour in deformed aluminium were observed. Type I behaviour occurs at lower temperatures and it is characterized by sharpening of the dislocation cell walls without any increase in the average cell size, while in type II recovery, at higher temperatures, a definite increase in cell size was reported. It could be speculated that the observed increase in the average squared derivatives at lower annealing temperatures is connected to type I recovery and the subsequent decrease to type II behaviour. It can be demonstrated, by using variational calculus, that of all functions \( e(z) \) giving the same value for \( \langle e(z)^2 \rangle \), a purely sine-shaped function gives a minimal value of \( \langle e'(z)^2 \rangle \). Any deviation from this 'smooth' behaviour that sharpens the gradients in parts of the region and correspondingly weakens the variation in others, will increase the value of the average squared derivative. The same is naturally also true for the average squared second derivative, the one for the first being kept constant, etc. Consequently, the sharpening of the cell boundaries should have an increasing effect on the averages of the squared strain derivatives. The decrease at the higher annealing temperatures could be attributed to the annihilation and movement of cell walls occurring during type II recovery. Then the average amplitudes of the variation in the strain are decreasing, and this counteracts the effect of the sharpening of the gradients. Recently, Wang Yuming, Lee Shansan & Lee Yenchin (1982) have reported a study of X-ray diffraction line broadening in deformed and annealed aluminium, but their interpretation is so different from the present one that comparison is not possible.

The strain field that is related to the present type of X-ray diffraction results arises mainly from the dislocations contained in the crystals. The strain field is a function of the density of the dislocations, and in particular of their exact arrangement. Therefore, for example, on the basis of models, it should be possible to find a direct connection between the dislocation density and arrangement and the averages of the squared strain and the squared derivatives as they are obtained from the X-ray measurements. The exact knowledge of such a connection would indeed be very useful in investigating various dislocation-related phenomena in materials. Even if no direct connection could be found on theoretical grounds, it is to be expected that different types of dislocation arrangements would give different relative values for the average squared derivatives. Some preliminary measurements carried out by the present authors seem indeed to indicate that this is the case. Thus, the relative values of the average squared derivatives of the strain could serve as 'finger prints' for different types of dislocation configurations.

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