**A. Segmüller**

# Determination of Lattice Strain and Crystallite Size in Thin Films

Warren and Averbach[1] have described a method for the determination of inhomogeneous lattice strain and crystallite size by means of the Fourier analysis of the x-ray line profile. Their technique requires the measurement of more than the first order of a reflection—in special cases of more than one line. In analyzing this technique, Wagner[2] has stated that it is in fact impossible to separate the influence of strain and crystallite size on the line profile by measuring only one line. However, if the sample is very thin, only one order of reflection and often only one line can be measured. For instance, for a nickel film of about 1000 A thickness and for copper $K_\alpha$-radiation, the integrated intensity of the (222) reflection is only about 4% of that of the (111). This considerable decrease of intensity is due to the Lorentz and polarization factor as well as the atomic scattering and absorption factor.

Smith[3] has described the determination of rms strain and average crystallite size in electroplated thin Permalloy films from the measurement of only one line. His technique is based on the following expression for the low-order Fourier coefficients of the x-ray line profile:

$$A_n = (1 - n/N_{av})\cos(2\pi n\varepsilon_{av})\,. \tag{1}$$

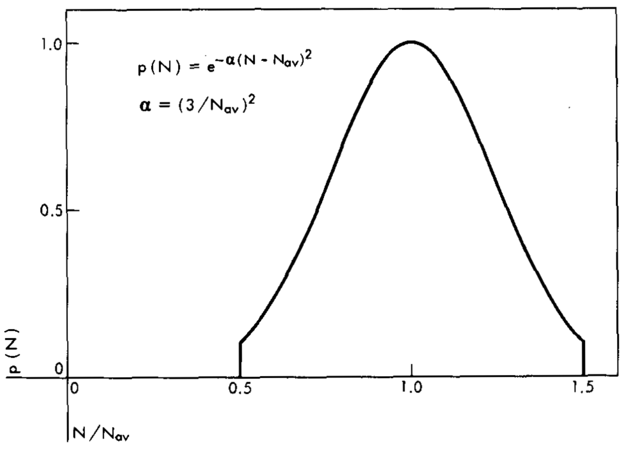
Here $N_{av} = D/a_0$, where $D$ is the average crystallite size and $a_0$ the distance of lattice planes, (both measured in the direction perpendicular to the reflecting lattice planes) and $\varepsilon_{av}$ is the rms strain in this direction. By choosing suitable values of $N_{av}$ and $\varepsilon_{av}$, it is possible to approximate the Fourier coefficients of the measured line profile with the theoretical curve of Eq. (1) for low values of $n$.

In measuring line profiles of thin films, however, there arise some difficulties which make this technique less useful in practice than would appear from the theoretical equation. The background noise is normally so high that considerable error is introduced in determining the width of the line profile at its base. Normally the background level is taken to be too high, and in the case of pure crystallite size broadening, Bertaut[4] has shown that this effect causes a negative curvature of the $A_n$-vs-$n$ curve for low $n$ instead of a constant slope, as is to be expected from Eq. (1) for $\varepsilon_{av} = 0$. This negative curvature (the so-called hook) would give a negative value of the size distribution function for very small crystals which is physically impossible.[2,4,5] Using Smith's method for such a curve results in a spurious rms strain, even for samples with no strain at all.

In order to estimate the error resulting from an erroneous determination of background intensity, a symmetrical line profile has been calculated for a given distribution (Fig. 1) and for zero strain. Following Bertaut's treatment, the calculated intensity curve $P(h)$, where $h$ is the coordinate in reciprocal space with $-1/2 \leqq h \leqq +1/2$, was cut at different positions $h_0 < 1/2$ of the line tail. The line profiles cut in this manner have the defining relations, $P'(h) = P(h) - P(h_0)$ for $h < h_0$ and $P'(h) = 0$ for $h \geqq h_0$. The Fourier coefficients for these profiles were computed and approximated by Eq. (1) in order to determine the spurious rms strain $\varepsilon_{av,0}$ and the corresponding crystallite size $N_{av,0}$ which might be different from $N_{av}$.

In Fig. 2 the cosine Fourier coefficients $A_n$ are plotted versus $n/N_{av}$. If one uses $h_0N_{av}$ as parameter,

*Figure 1* **Crystal size distribution function cut off for convenience of computing, and chosen broad enough to smooth out the secondary maxima of line profile.**



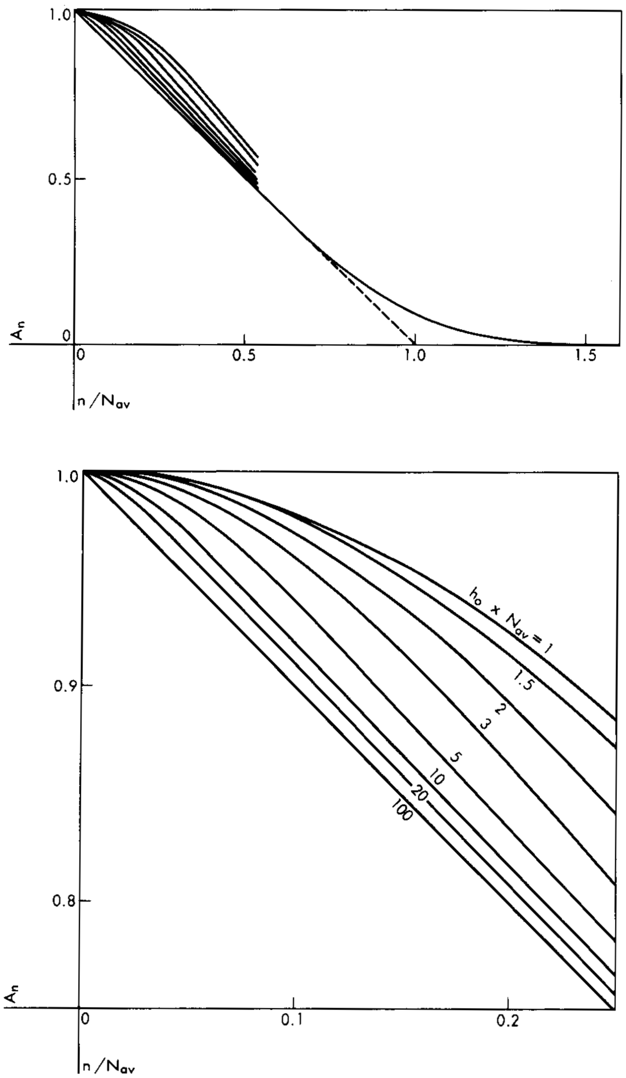
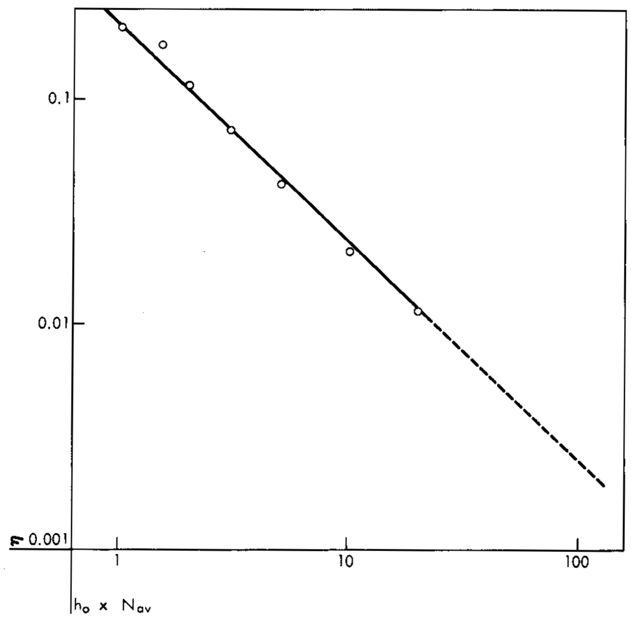



*Figure 2* **Fourier coefficients $A_n$ of cut line profiles normalized for $A_0 = 1$.**
*The $A_n$ of the original line profile, corresponding to the size distribution of Fig. 1 and to zero strain, are given by the lowest curve, the straight part of which is extrapolated by the dotted curve towards $A_n = 0$ at $n/N_{av} = 1$. Below at left, $A_n$ are shown in enlarged scale with the cutting parameter $h_0 \cdot N_{av}$.*

*Figure 3* **Error $\eta$ of the zero-order Fourier coefficient $A_0$ due to cutting the line profile at $h_0$.**




the curves $A_n$ versus $n/N_{av}$ agree very closely for different values of $N_{av}$. This correspondence can be shown to result from the dependence on $hN$ of the numerator in the formula for the intensity curve

$$P(h) = \sin^2(\pi hN)/\sin^2(\pi h) \,. \tag{2}$$

Figure 3 shows the error $\eta$ of the zero-order Fourier coefficient $A_0$—the area enclosed by the line profile—caused by cutting the intensity curve at $h_0$. If one divides the Fourier coefficients for $n \geqq 1$ by $1 + \eta$, they agree very well with the original ones except for low values of $n$, since the low orders are also influenced by the cutting procedure. From Fig. 3 the following very simple approximation for $\eta$ can be obtained:

$$\eta = 0.2/(h_0 N_{av}) \,. \tag{3}$$

Figure 4 shows the crystallite size $N_{av,0}$ and the spurious strain $\varepsilon_{av,0}$ as determined by fitting the Fourier coefficients of Fig. 2 with Eq. (1) for $n < 0.5 \cdot N_{av}$. The scale for $\varepsilon_{av,0}$ holds only for $N_{av} = 200$. For other values of $N_{av}$ one has to multiply the values of Fig. 4 by $200/N_{av}$ to obtain the spurious rms strain $\varepsilon_{av,0}$.

For practical purposes, a measuring interval five to ten times as large as the halfpeak breadth $2h_{0.5}$ seems fairly reasonable. Since for a line profile without strain broadening, the halfline breadth is given by $h_{0.5} = 0.4/N_{av}$, typical values of $h_0 N_{av}$ range from 2 to 4, causing an error of the determined crystallite size between 50 and 150% and an erroneous strain of about $(200/N_{av}) \cdot 0.1\%$. This error might be increased or decreased by the statistical uncertainty of background determination. The resulting error of $A_0$ is given by

$$\Delta A_0 = \eta A_0 \pm 2\Delta I_b \cdot h_0 \,, \tag{4}$$

where $\Delta I_b$ is the statistical error of the background intensity. Only if the second term on the right side of (4) is small compared to the first one, a correction of $A_0$ can be performed with Eq. (4). However, it should be considered as an approximation because $\eta$ is

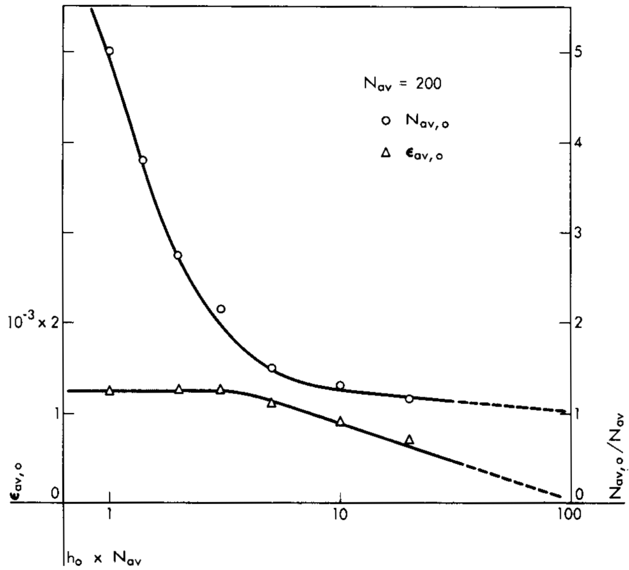



*Figure 4* **Spurious rms strain $\varepsilon_{av,0}$ (for $N_{av} = 200$) and corresponding crystallite size $N_{av,0}$ caused by cutting the line profile at $h_0$.**

defined only for strain-free samples by Eq. (3).

Another source of error is mentioned by Warren,[5] namely, the fact that the line profile never reaches the true background level, even at $h = 1/2$. However, the resulting error of $A_0$ amounts only to about 1% for $N_{av} = 40$ and, therefore, it can be ignored for crystallite sizes $N_{av} > 40$.

A good fit of the experimental Fourier coefficients by the theoretical formula (1) with suitably chosen parameters[3] $N_{av}$ and $\varepsilon_{av}$ should not be considered as evidence for correct background determination, because it is easy to fit curves fairly well with a hook, e.g., the curves of Fig. 2.

From these considerations the following conclusions can be drawn. In most cases a hook appears in the $A_n$-vs-$n$ curve for low values of $n$. The form of this hook is very sensitive to the background level assumed for the evaluation of the Fourier coefficients. If there is no strain in the sample, e.g., in a very fine powder, then this hook is normally corrected by extrapolating the straight part of the $A_n$-vs-$n$ curve towards $n = 0$ and it is possible to compare this correction with Eq. (3). In the presence of lattice strain, as generally occurs in thin films, the possibility of a correction with Eq. (3) depends on the uncertainty of the background level.

## References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