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X-ray Diffraction Line Broadening Due to Dislocations in Non-Cubic Crystalline Materials. III. Experimental Results for Plastically Deformed Zirconium

BY RADOMÍR KUŽEL JR

Department of Semiconductor Physics, Faculty of Mathematics and Physics, Charles University, 121 16 Prague 2, Czechoslovakia

AND PETER KLIMANEK

Department of Materials Science, Academy of Mining, 9200 Freiberg, German Democratic Republic

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Dedicated to the memory of Professor Dr M. A. Krivoglaz

Abstract

Procedures of X-ray diffraction line profile analysis for the evaluation of the dislocation content in plastically deformed hexagonal materials were tested by means of conventional powder diffractometry on polycrystalline zirconium deformed under tension at 77 K. In order to obtain a representative picture of the dislocation-induced X-ray line broadening a series of reflections was measured. The integral breadths and the Fourier coefficients were evaluated by both direct profile-shape analysis and profile fitting with analytical functions. The results show a significant anisotropy of the line broadening. The 000*l* reflections are clearly less broadened than most of the others. According to the theoretical calculations presented previously such a phenomenon can be expected if the plastic deformation favours generation of dislocations with Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$.

1. Introduction

In the first and second papers of this series (Klimanek & Kužel, 1988; Kužel & Klimanek, 1988; henceforth papers I and II) the orientation factors controlling the anisotropy of dislocation-induced X-ray line broadening were calculated for polycrystalline hexagonal materials. The main features of this anisotropy are determined by the relative content of dislocations with Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$ and $a/3\langle 11\bar{2}3\rangle$. In comparison with those of the other diffraction peaks the former leads to very small and the latter to very large broadening of the 000*l* reflections.

In order to find some experimental indication of the effect just mentioned the literature on XRD line-profile analysis of hexagonal materials was searched. A dominant content of dislocations with Burgers vector $a/3\langle 2\bar{1}\bar{1}0\rangle$ can be expected for many h.c.p. metals especially high-melting ones (Ti, Zr; Predvoditelev & Troickyi, 1973), and consequently a low broadening of the 000*l* reflections should be typical for them. Indeed in some measurements (*e.g.* Mogard & Averbach, 1958; Lele & Anantharaman, 1967; De & Sen, 1968; Bengtsson, Johannesson & Lindau, 1971) on powder specimens the phenomenon is visible. A reduced strain broadening of the 000*l* lines has also been observed for zinc films (Suchitra Sen, Nandi & Sen Gupta, 1978). However, from other results on zirconium filings (Chatterjee & Sen Gupta, 1974), zirconium deformed in compression (Byugov, Samsonik & Sirenko, 1976), titanium filings (Metzbower, 1971) or titanium cold-worked in various modes (Raychenko, Martynova, Kononenko, Geletiy & Tuzyiak, 1969), no unambiguous conclusions concerning the line-broadening anisotropy can be drawn, and in many cases the presentation of the results is too incomplete for this purpose.

Dislocations with Burgers vectors $a/3\langle 11\bar{2}3\rangle$, which should lead to very broad 000*l* reflections, are probable, above all, in low-melting h.c.p. metals such as, for instance, zinc and cadmium (*e.g.* Predvoditelev & Troickyi, 1973). But unfortunately the line-broadening anisotropy can hardly be measured for them at room temperature, since the strain broadening of the X-ray diffraction peaks is drastically reduced by recovery and recrystallization even in filings (Halder & Hunter, 1974; Halder & Johnston, 1975).

Because of the difficulties just mentioned a more detailed investigation of the line broadening in hexagonal materials was performed with polycrystalline zirconium after tensile deformation at 77 K. The effect was studied by both the integral breadth and the Fourier coefficients of a representative number of reflections. However, the aim of the paper is to demonstrate the application of the formalism and conclusions described in papers I and II and not to study the plastic deformation of zirconium.

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2. Experimental procedures and methods of evaluation

2.1. *Specimen treatment and diffraction experiments*

The sample object of the present work was a flat tensile specimen of polycrystalline zirconium with 99·99% purity and a mean grain size of 30 μm, which was plastically deformed by tension up to an elongation of 12% at 77 K (in liquid nitrogen). The low deformation temperature was chosen in order to avoid deformation of the dislocation arrangement with significant defect correlation (*e.g.* a dislocation cell structure), which is typical for deformation processes above 293 K (*e.g.* Dlouhý, 1986).

X-ray diffraction patterns of the material were measured with a Philips PW1380 powder diffractometer, using Cu $K\alpha$ radiation of an X-ray tube with normal focus (40 kV, 30 mA), Soller slits and a graphite monochromator within the diffracted beam and a proportional counter as detector. All diffraction peaks were measured with θ–2θ step scanning.

In order to check the reliability of the line profiles three sets of reflections were measured in the specimen plane perpendicular to the axis of tension. The first experiment (I, specimen centre, 11 reflections) took place immediately after the deformation, the second one (II, specimen centre, 11 reflections) after removal of a surface layer of about 30 μm thickness by etching and the third one (III, away from the specimen centre, 21 reflections) after several months. The irradiated area was about 25 mm^2.

2.2. *Evaluation of the diffraction line profiles*

The parameters of the XRD line profiles were determined from the measured intensity distributions either directly or after fitting with analytical functions. For this purpose two computer programs were written in Fortran: *PROAN* for the direct analysis and *PROF* for the fitting of the line profiles. *PROAN* is suitable for isolated peaks, if the background on both sides of the profile can be well estimated. It includes the following steps: (1) background correction; (2) correction for the angle-dependent intensity factors (Lorentz, polarization), conversion to sin θ scale; (3) doublet separation (Rachinger method); (4) determination of profile characteristics (peak position, integrated intensity, integral breadth, half width – FWHM, moments); and (5) Fourier analysis.

The second program *PROF*, the general structure of which is very similar to that of *PROAN*, is an extended version of a procedure presented by Valvoda & Voland (1981). It allows the treatment of line groups of an XRD pattern, and up to eight peaks can be separated. It is based on the Levenberg–Marquardt least-squares method (Marquardt, 1963) and offers several functions (Pearson, pseudo-Voigt and rational; see Table 2 for references) in symmetrical and asymmetrical versions. The doublet separation and/or the background correction can be included in the fitting procedure. From the fitted profiles the diffraction line parameters and the Fourier transforms are obtained by numerical integration with a significant reduction of the truncation effects. In the present work the Pearson VII or the pseudo-Voigt function was a sufficient approximation for the diffraction line profiles, and the GOF factor (goodness of fit; *cf.* Young & Wiles, 1982) was within the range 1–3·5% in most cases. The application of *PROF* may be illustrated by Fig. 1. Values of the integral breadths, which were directly determined from the measured line profiles by *PROAN*, are given in Table 1 for all the experiments.

2.3. *Correction of the instrumental line broadening*

In order to find the instrumental line broadening of the measured diffraction peaks, the annealed zirconium sample was used as a standard before the plastic deformation. The line broadening of its reflections was controlled by measurements with a well tested powder standard of WC, and within the limits of the experimental accuracy good agreement of the results was obtained. The peaks of the plastically deformed zirconium sample were at least twice as broad as the profiles of the standard.

The contribution of the instrumental line broadening to the Fourier coefficients of the diffraction peaks were eliminated in the usual manner by means of the Stokes correction (Stokes, 1948; *cf.* Klug & Alexander, 1974, for instance). For a direct correction of the integral breadths the Voigt-function method described by de Keijser, Langford, Mittemeijer & Vogels (1982) was used. The reliability of the procedure was controlled by comparison with line widths, which were calculated from the Stokes-corrected Fourier coefficients, and was found to be very good (Table 2). According to work of Klimanek

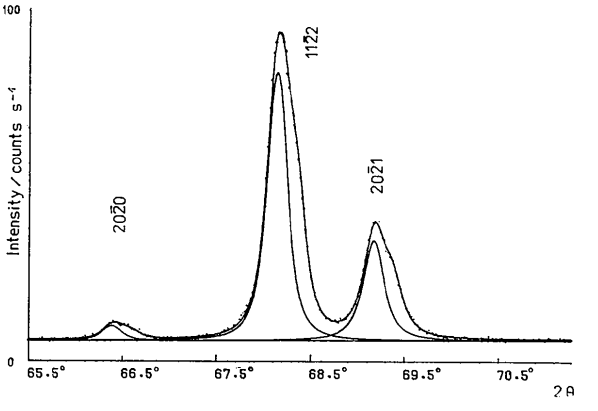

Fig. 1. A segment of the diffraction pattern. The dotted data correspond to the experimental intensities (counts s^{-1}) corrected for the Lorentz and polarization factors, the upper solid line to the total fitted function and the lower solid lines to the α_1 peaks of individual reflections.

Table 1. *Experimental values of integral breadth for three measurements on zirconium deformed at* 77 K [*in* 10^{-3} Å *of the* $2(\sin\theta)/\lambda$ *scale*]

Reflection	I	II	III
$10\bar{1}2$	3·16	2·98	2·41
$11\bar{2}0$	3·09	3·30	2·82
$10\bar{1}3$	3·63	3·07	2·91
0004	3·58	2·95	2·70
$20\bar{2}2$	4·09	4·65	3·48
$10\bar{1}4$	4·18	3·73	3·44
$20\bar{2}3$	4·28	3·75	3·75
$11\bar{2}4$	4·18	4·19	3·98
$21\bar{3}3$	5·14	4·76	4·40
$30\bar{3}2$	4·82	4·80	4·40
0006	4·72	4·17	4·08

Table 2. *Integral breadth* [*in* 10^{-3} Å *of the* $2(\sin\theta)/\lambda$ *scale, measurement* II] *corrected for instrumental effects by means of the method of Voigt function* (*Voigt*) *and the Stokes method through Fourier coefficients* (*Fourier*)

Uncorrected values were determined by the direct line-profile analysis with analytical doublet separation (D) and by profile fitting with variable Lorentzian (vL), pseudo-Voigtian (pV) and rational (R) functions

	Fourier				Voigt			
Peak	D	vL	pV	R	D	vL	pV	R
$10\bar{1}2$	2·15	2·09	2·11	2·00	2·03	2·10	2·11	2·06
$11\bar{2}0$	2·30	2·30	2·51	2·40	2·39	2·32	2·55	2·48
$10\bar{1}3$	2·40	2·32	2·34	2·35	2·35	2·47	2·46	2·35
0004	2·36	2·25	2·28	2·27	2·09	2·05	2·22	2·09
$20\bar{2}2$	3·16	2·86	3·01	2·62	2·85	2·76	3·12	2·85
$10\bar{1}4$	2·76	2·81	2·84	2·84	2·66	2·57	2·92	2·66
$20\bar{2}3$	3·06	2·97	2·94	3·01	3·06	2·89	2·88	2·88
$11\bar{2}4$	3·05	3·07	3·04	3·07	3·24	3·20	3·10	3·10
$21\bar{3}3$	4·08	3·92	3·90	3·96	3·96	3·92	3·95	3·96
$30\bar{3}2$	4·13	4·04	4·03	3·96	4·08	3·95	3·94	3·94
0006	3·19	3·11	3·16	3·24	3·07	3·03	3·07	3·07

Functions with parameters A_i:

(1) variable Lorentz $y = A_1/[1 + A_3(x - A_2)^2]^{A_4}$ similar to Pearson VII;

(2) pseudo-Voigt $y = A_1\{A_4/[1 + A_3(x - A_2)^2] + (1 - A_4)\exp[-\pi^2 A_3(x - A_2)^2]\}$;

(3) rational $y = A_1/[1 + A_3(x - A_2)^2 + A_4(x - A_2)^4]$.

References: Hall, Veeraraghavan, Rubin & Winchell (1977) for (1); Enzo, Polizzi & Benedetti (1985) for (2); Pyrros & Hubbard (1983) for (3).

(1988*a*, *b*) this also indicated that the structural inhomogeneity (fluctuations of the defect content in different grains) of the specimen was sufficiently small.

The shapes of the diffraction lines were estimated by comparison of the double normalized Fourier coefficients of the measured profiles with the Fourier transforms of standard functions (Oettel, 1971). They were found to be effectively close to the quadratic Cauchy function.

2.4. *Determination of the dislocation content*

Fourier coefficients. According to Krivoglaz (1983) the cosine Fourier coefficients $A(L)$ of a diffraction line profile which is broadened due to an ensemble of dislocations with moderate defect correlation [a more general description of dislocation-induced X-ray line broadening can be found in recent work of Groma, Ungár & Wilkens, (1988)] and the same type of Burgers vector **b** are given by the expression

$$\ln A(L) = -BL^2 \ln (r_c/L) \qquad (1)$$

with

$$B = \chi\rho b^2 Q^2/8\pi;$$

$Q = (4\pi/\lambda)\sin\theta$ is the magnitude of the diffraction vector **Q**. The parameter r_c is a factor connected with the outer cut-off radius of the dislocations and characterizes the defect correlation, the quantity ρ denotes the (total) dislocation density, and χ is the orientation factor as defined in papers I and II.

As follows from (1) and the limitations of its applicability (Krivoglaz, 1983) a plot of $\ln A(L)/L^2$ *vs* $\ln L$ should be linear for low and middle values of the distance L. (A plot of this kind, typical for all the reflections of the present work, is shown in Fig. 2.) Accordingly, the slope of its linear part gives the value of B and the intercept with the $\ln L$ axis is $\ln (r_c)$. For practical conditions the application of the procedure requires that no line broadening due to particle size or stacking faults contributes to the Fourier coefficients. Otherwise the Warren–Averbach analysis (*e.g.* Warren, 1969) has formally to be used for the separation of the dislocation-induced strain broadening. The mean square strains $\langle\varepsilon^2(L)\rangle$ are then related to the dislocation density by the formula

$$\langle\varepsilon^2(L)\rangle = (1/4\pi)\chi\rho b^2 \ln (r_c/L). \qquad (2)$$

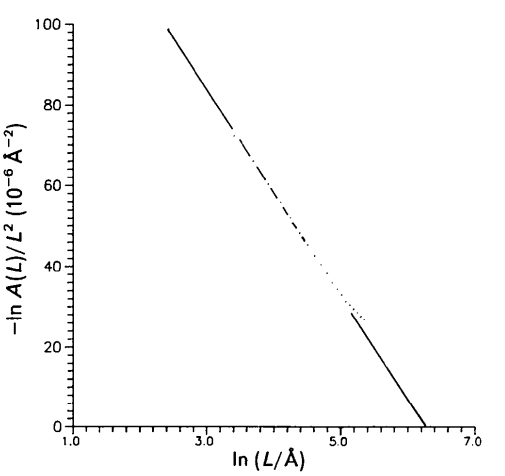

Fig. 2. A typical example of the dependence of the logarithm of Fourier coefficients over L^2 *vs* $\ln L$ [equation (1)] with a substantial linear part.

Hence, from the linear part of the dependence of $\langle\varepsilon^2(L)\rangle$ *vs* ln L both the quantity B/Q and the correlation parameter r_c can be determined.

In order to estimate the total dislocation density ρ of the scattering material from experimentally determined parameters B, the orientation factors must be known. This requires at least some qualitative knowledge (or assumptions) concerning the possible dislocations occurring in the structure considered (Burgers vectors, dislocation character, slip planes). If only one family of Burgers vectors or slip planes has to be taken into account, as, for example, in cubic metals and alloys, the calculation of χ is relatively easy and can, in principle, be obtained from the line broadening of a single reflection. In the case of hexagonal (or other non-cubic) materials with different types of Burgers vectors and/or slip systems a set of reflections is necessary for this purpose and the following procedure can be applied: Corresponding to (1) a plot $B_\chi = (B/\chi)$ *vs* $\sin^2\theta$ with the correct values of the orientation factors must be a straight line, the slope of which is proportional to the dislocation density. This means that it is necessary to determine the true factors χ on the basis of a successive elimination of the observed line-broadening anisotropy by appropriate combination of the orientation factors calculated for different fractions of the expected dislocation types.

If a number of reflections can be measured, a tempting idea for a more detailed analysis of the dislocation content may be the procedure of 'dislocation content fitting'. In a first approximation a vector of integral breadths can be used for this purpose, which assumes the factor ln r_c to be independent of the direction of the diffraction vector **Q** and has to be constructed as a linear combination

$$(B\lambda^2/\sin^2\theta) = 2\pi\rho \sum_{i=1}^{\nu} c_i b_i^2 \chi_i(hkl) = B'(hkl) \qquad (3)$$

of the vectors $\mathbf{B}'$ of the orientation factors associated with different fractions of the possible dislocation types. Since the quantities $\chi_i = \chi_i(hkl)$, $\mathbf{B}' = B'(hkl)$ are n-dimensional vectors (n is the number of reflections), the desired coefficients c_i could be obtained by a least-squares method, provided $\nu < n$. Of course, the practical application of the procedure assumes experimental data of very high accuracy.

Integral breadths. The integral breadth of an X-ray reflection is relatively insensitive to experimental errors and can easily be calculated. However, the determination of the dislocation density is not so straightforward as in the case of Fourier coefficients. According to Krivoglaz (1983) the integral breadth of a diffraction line profile related to a dislocation arrangement with weak defect correlation is given by the relation

$$\beta(2\theta) = 4e \tan\theta \qquad (4a)$$

or in units of $2(\sin\theta)/\lambda$ as

$$\beta = 4e(\sin\theta)/\lambda \qquad (4b)$$

with

$$e^2 = \chi\rho b^2 A^2 \ln P/8$$

and

$$P = rB^{1/2}; \quad A = \{1 - \ln(\ln P)/4\ln P\}^{-1}. \qquad (5)$$

The correct way for determining the correlation parameter P is to estimate it experimentally either from the Fourier coefficients (see above) or from the shape of the physical diffraction line profile (Wilkens, 1970). The application of (4) is entirely justified for values of $\ln P > 2$ (Krivoglaz, 1983), but according to numerical estimates of the line breadth from (1) it can also be used as an approximation for $\ln P > 1$ (Kužel, 1987). In this connection it has to be noticed that the uncertainties in the application of (4) cause only a systematic error of the absolute values of the dislocation densities, but they do not change the procedure for the analysis of the line-broadening anisotropy.

If no particle-size effect has to be taken into consideration, the dislocation density of a non-cubic material containing dislocations with different types of Burgers vectors can, again taking advantage of the line-broadening anisotropy, be estimated directly by means of a set of reflections from a plot of β_χ *vs* $\sin\theta$. As in the case of the Fourier coefficients such a plot must become a straight line if the factors are correctly chosen.

3. Results and discussion

3.1. *Dislocation characteristics and orientation factors of zirconium*

In order to calculate the orientation factors of dislocation-induced diffraction line broadening, the literature dealing with transmission electron microscopy (TEM) of plastically deformed zirconium was studied. It was found that especially representative data concerning the relations between the dislocation density and the deformation conditions are very rare.

The Burgers vectors (and slip systems) of the dislocations occurring in cold-worked zirconium were studied by several authors (Bailey, 1962; Westlake, 1965; Bedford & Miller, 1971; Tenckhoff, 1972; Akhtar, 1973; Northwood & Gilbert, 1973; Woo, Carpenter & MacEwen, 1979; Ballinger, Lucas & Pelloux, 1984; Dlouhý, 1986; Merle, 1987). The results can be summarized as follows:

(a) Slip deformation is mainly caused by $\langle a\rangle$ dislocations with Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$ on prismatic

slip planes, but particularly in polycrystals such dislocations are also observed on basal and pyramidal slip planes. After low-temperature deformation [100 K (Westlake, 1965)] an increased contribution of the basal slip was observed.

(*b*) Because of their higher energy the fraction of $\langle a+c\rangle$ dislocations with Burgers vector $a/3\,\langle 11\bar{2}3\rangle$ is generally small. The probability of their occurrence increases at higher stresses and/or temperatures and especially in deformation conditions which are unfavourable for pure $\langle a\rangle$ slip. Obviously they are also necessary for the accommodation of strain in different grains of a polycrystal and therefore are preferentially observed near twins, grain boundaries and within very small grains (Woo, Carpenter & MacEwen, 1979). Moreover, they can be expected in dislocation arrangements with high defect correlation [cell structures, networks, sub-boundaries (*cf.* Northwood & Gilbert, 1973; Dlouhý, 1986)].

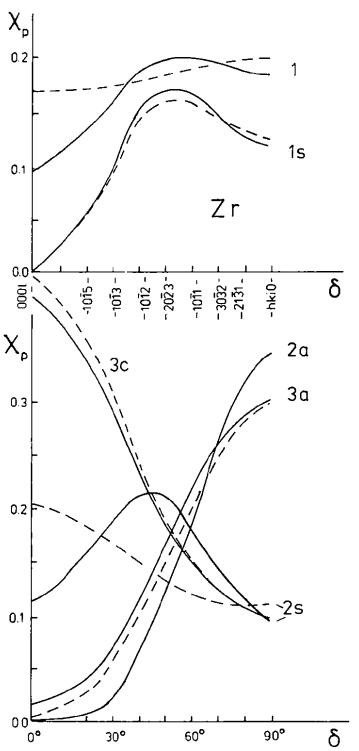

Fig. 3. The dependence of orientation factors χ_p on the angle between the reciprocal-lattice vector connected with the operating diffraction vector and the *c* axis of the hexagonal lattice. Calculations were performed for the following dislocations (the notation is the same as in papers I and II):

screw	1*s*	$\langle 2\bar{1}\bar{1}0\rangle$, 2*s* $\langle 11\bar{2}3\rangle$
edge	1	basal $\langle 2\bar{1}\bar{1}0\rangle$
	2*a*	prismatic $\langle 2\bar{1}\bar{1}0\rangle$
	3*a*	pyramidal $\langle 2\bar{1}\bar{1}0\rangle$ $\{0\bar{1}11\}$
	3*c*	pyramidal $\langle\bar{2}113\rangle$ $\{2\bar{1}\bar{1}2\}$
	3*b*	pyramidal $\langle\bar{2}113\rangle$ $\{10\bar{1}1\}$ (not shown; similar to 3*c*).

The dashed lines correspond to the calculations in the approximation of elastic isotropy.

(*c*) Another important deformation mechanism of polycrystalline Zr at low temperatures is mechanical twinning (*e.g.* Reed-Hill, Buchanan & Caldwell, 1965; Baldwin & Reed-Hill, 1968; Flade, 1971; Akhtar, 1973). Its intensity is strongly influenced by the deformation mode (compression is more favourable), the deformation temperature, the texture of the specimen and the impurity content in the material. As found by Flade (1971) and Dlouhý (1986) in agreement with the cited literature, in tensile tests at 77 K the effect of twinning is small up to strains $\varepsilon = 0{\cdot}15$, and the thickness of the twin lamellae is relatively large. For this reason a significant influence of twinning on the diffraction-line broadening observed in the present work can be excluded.

According to the results of the TEM studies the orientation factors of zirconium were calculated for various $\langle a\rangle$ and $\langle a+c\rangle$ dislocations. The results are plotted in Fig. 3 as a function of the angle between the *c* axis of the hexagonal lattice and the diffraction vector (see paper I) for both the approximation of elastic isotropy (dashed lines) and the general case of elastic anisotropy. The main features of the results are similar to those for magnesium (paper I), and it is therefore not necessary to discuss them here. The averaged orientation factors $\langle\chi_p\rangle$ of Fig. 3 were calculated for the case of elastic anisotropy, assuming various fractions of the $\langle a\rangle$ and $\langle a+c\rangle$ dislocations. (The corresponding results obtained in the approximation of elastic isotropy have already been given in Fig. 5 of paper I.*) The following modelling assumptions concerning the dislocation arrangement were used:

(i) content of $\langle a\rangle$ dislocations: 50% screws and 50% edges (33·33% of them on slip systems $\langle 2\bar{1}\bar{1}0\rangle\{0001\}$, 33·33% on $\langle 2\bar{1}\bar{1}0\rangle\{01\bar{1}0\}$ and 33·33% on $\langle 2\bar{1}\bar{1}0\rangle\{0\bar{1}11\}$);

(ii) Content of $\langle a+c\rangle$ dislocations: 50% screws and 50% edges (50% of them on $\langle 11\bar{2}3\rangle\{0\bar{1}11\}$, 50% on $\langle 11\bar{2}3\rangle\{\bar{1}\bar{1}22\}$).

A generalized treatment only slightly modifies the shape of the curves of $\langle\chi_p\rangle$ *vs* δ.

3.2. *X-ray determination of the dislocation with various Burgers-vector content*

In order to interpret the physical line broadening of plastically deformed zirconium the corrected integral breadths and the quantities *B* obtained from the evaluation of the Fourier coefficients were plotted in the manner proposed in § 2.4. The results are illustrated by plots for experiment III in Fig. 4 (line widths) and Fig. 5 (Fourier coefficients). The zero intercept of both lines with the ordinate supports the assumption of pure strain broadening of the X-ray

* The axis labels of Fig. 5 in paper I and Fig. 3 in paper II are incomplete. The quantities actually plotted are $(6^{1/2}/a)\langle\chi_p\rangle^{1/2}$ and $(6/a^2)\langle\chi_p\rangle$, respectively, and not $\langle\chi_p\rangle^{1/2}$ and $\langle\chi_p\rangle$.

reflections. Therefore a straight line can be fitted to the values, the slope of which leads to the dislocation density. The error bars of the data characterize both the uncertainties arising from the counting statistics and the differences between the results of various evaluation procedures (*cf.* Table 2). The spread of values of B (Fig. 5) is greater than that of the line breadths (Fig. 4) because the determination of B from the Fourier coefficients is relatively sensitive to systematic errors due to the profile fitting, the correction of the instrumental line broadening *etc.* However, in both cases an anisotropy of the line broadening can clearly be detected, which corresponds to the predictions of papers I and II and, moreover, to TEM results on the dislocation structure of zirconium. The $000l$ peaks are significantly less broadened than most of the others, which indicates a dominating content of dislocations with the Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$ ($\langle a\rangle$ dislocations).

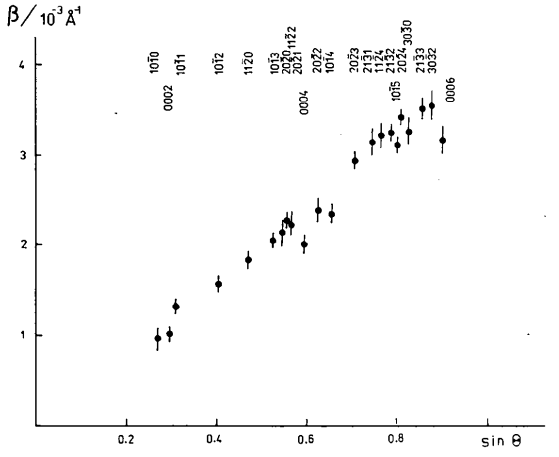

Fig. 4. Dependence of integral breadths β [$(2\sin\theta)/\lambda$ scale] on $\sin\theta$ for measurement III.

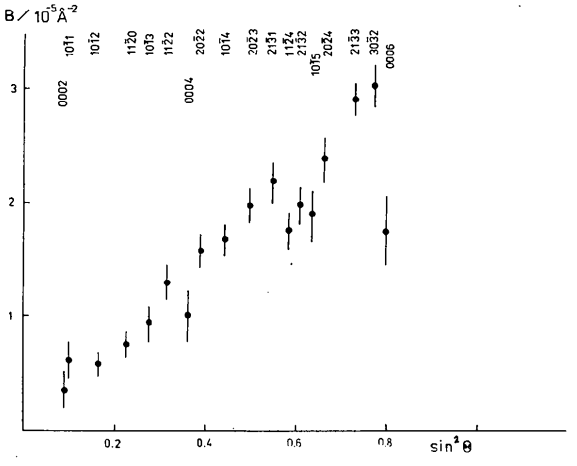

Fig. 5. Dependence of the quantity B determined from Fourier coefficients on $\sin^2\theta$ [equation (1)] for measurement III.

The elimination of the anisotropy in the experimentally determined line widths and B parameters by means of suitably averaged orientation factors (see § 3.1) is demonstrated in terms of quantities $\beta/\langle\chi_p\rangle$ (line widths) in Fig. 6 and $B/\langle\chi_p\rangle$ (Fourier coefficients) in Fig. 7. In all cases a minimal spread of values (black dots) around a straight line was found for 80% of the dislocations with Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$ and 20% of the dislocations with Burgers vectors $a/3\langle 11\bar{2}3\rangle$. In a more refined analysis the ratio of the $\langle a\rangle$ and $\langle a+c\rangle$ dislocations seemed to be shifted towards the values 85:15 (measurement III - Figs. 6, 7) or even 90:10 (measurements I, II). From Figs. 6 and 7 it also follows that the $000l$ peaks are very sensitive to the dislocation content as predicted in the preceding papers I and II.

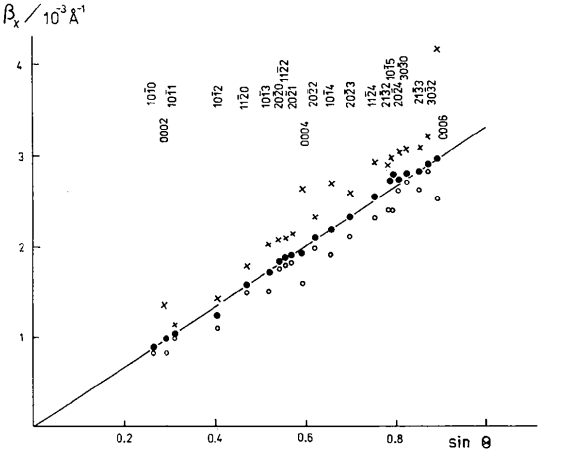

Fig. 6. Integral breadths corrected for the orientation factors $\langle\chi_p\rangle$ and plotted *versus* $\sin\theta$. The symbols correspond to simulation of different fractions of dislocations with Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$ and $a/3\langle 11\bar{2}3\rangle$, respectively (× 90:10, ● 80:20, ○ 70:30%). The results are shown for measurement III. The units are the same as in Fig. 4.

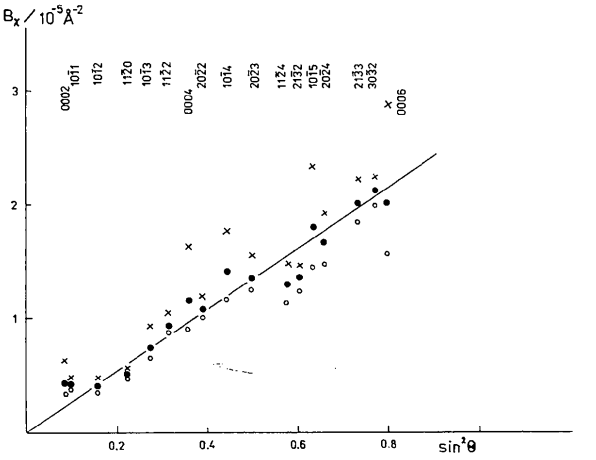

Fig. 7. The B quantities corrected for the orientation factors in a similar manner as for integral breadth in Fig. 6.

The anisotropy of the diffraction-line broadening in hexagonal materials is mainly determined by the relative content of $\langle a \rangle$ and $\langle a+c \rangle$ dislocations, but by investigation of special reflections it should be possible to describe the dislocation content in a more sophisticated manner, *i.e.* to distinguish various (screws or edges in different slip systems) $\langle a \rangle$ and/or $\langle a+c \rangle$ dislocations. The procedure is based on a careful analysis of the differences of the line broadening of peaks such as, for example 0002, 0004, 0006 and $10\bar{1}0$, $10\bar{1}1$, $10\bar{1}2$, $10\bar{1}3$ *etc.*, and can be illustrated by Fig. 8. There the quantities $\beta/\langle\chi_p\rangle$ of the reflections measured in experiment III are plotted against $\sin\theta$ after division by differently averaged orientation factors. The example illustrates how the mixing of different types of dislocations with a single kind of Burgers vector would be reflected in experimentally-observed line broadening anisotropy. The quantities $\langle\chi_p\rangle$ were calculated for various fractions of screw and edge dislocations of prismatic and basal slip systems with Burgers vectors $a/3\langle 2\bar{1}\bar{1}0\rangle$. The primary assumption was that, as found in the evaluation of Figs. 6 and 7, the ratio of the $\langle a \rangle$ and $\langle a+c \rangle$ dislocations is 80:20, but then the fractions of different $\langle a \rangle$ dislocations were varied. The plots of Fig. 8 correspond to (*a*) pure basal slip, (*b*) pure prismatic slip, (*c*) only screw dislocations, and (*d*) only prismatic edge dislocations. Since only the spread of the data is of interest the plots are shifted along the ordinate axis.

From Fig. 5 it can be seen that the ratios of the line widths of *hki*0 reflections (*e.g.* $11\bar{2}0$) and of diffraction peaks due to planes inclined about 40–60° with respect to the 000*l* planes (*e.g.* $10\bar{1}2$) are sensitive to the relative content of edge and screw $\langle a \rangle$ dislocations. The lowest scatter, which, however, is greater than that observed for the combination of screw and edge dislocations on three different slip systems (§ 3.1, Figs. 6 and 7), is obtained for a mixture of screw and prismatic edge dislocations (Fig. 8*b*). In agreement with the TEM studies cited above this result indicates that the plastic deformation of polycrystalline zirconium does not take place by prismatic slip only. In this connection it is interesting that the fit can be improved if the 20% $\langle a+c \rangle$ dislocations are assumed as pure edges (Fig. 8*e*). But with respect to the integrating character and the inevitable uncertainties of the X-ray analysis of the dislocation content, the data presented in Fig. 8 should not be overestimated.

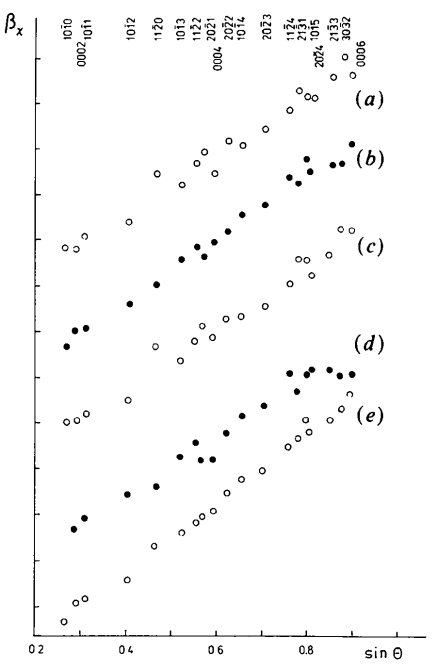

Fig. 8. Integral breadth corrected for the orientation factors and calculated for various simulations of the dislocation content. The fractions of $\langle a \rangle$ and $\langle a+c \rangle$ are always 80 and 20%, respectively, except for case (*d*) (70:30%). The contents of the $\langle a \rangle$ dislocations are as follows: (*a*), (*e*) basal slip (equal content of screw and edge dislocations); (*b*) prismatic slip (equal content of screws and edges); (*c*) only screw dislocations; (*d*) only prismatic edge dislocations ($\langle 2\bar{1}\bar{1}0\rangle\{0\bar{1}10\}$). The contents of the $\langle a+c \rangle$ dislocations are: (*a*)–(*d*) 50% of screw dislocations, 25% of the edge dislocations in the planes $\{0\bar{1}11\}$ and 25% in the planes $\{\bar{1}122\}$; (*e*) only edge dislocations in the planes $\{0\bar{1}11\}$.

3.3. *Dislocation densities*

The dislocation densities of the plastically deformed zirconium were estimated from: (*a*) the parameters *B* obtained from the Fourier coefficients by a plot of $\ln A(L)/L^2$ *vs* $\ln L$ (*cf.* Fig. 2); (*b*) the physical integral breadths β of the diffraction peaks; and (*c*) a formal evaluation of strain broadening by means of the Warren–Averbach analysis with the use of reflections with similar orientation factors (Fig. 9);

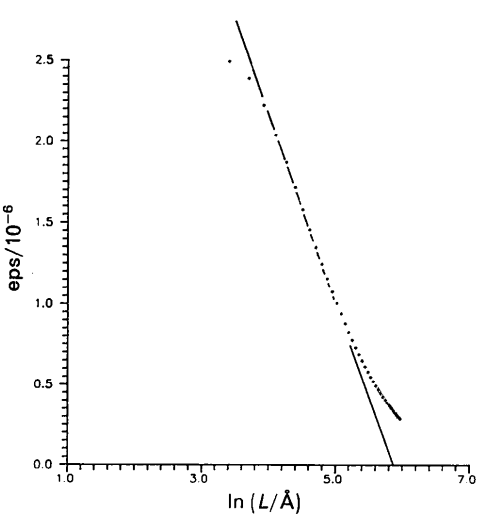

Fig. 9. Mean square strain $\langle\varepsilon^2\rangle$ as obtained from the Warren–Averbach method (see § 3) plotted against $\ln L$. The slope of the plot gives the dislocation density and the intercept gives the correlation parameter (modified cut-off radius).

Table 3. *Dislocation densities* (10^{14} m^{-2}) *determined from the dependence of integral breadths on* $\sin\theta$ (ρ_β), *from the dependence of the quantities B on* $\sin\theta$ (ρ_B) *and from the Warren–Averbach method* (ρ_ε)

The ranges of the P factors for high-angle reflections are also given.

Measurement	ρ_β	ρ_B	ρ_ε	P
I	4·3	6·2	5·5	4
II	3·9	5·2	5·3	5
III	3·5	6·0	5·6	5–6

after correction for the averaged orientation factors. For the analysis of the line breadths it was necessary to estimate the factors $P(5)$ from the Fourier coefficients. All values of P were within the range 3·5–7 which, according to the results of the profile fitting and in agreement with numerical calculations of line shapes by means of (1), corresponds to a distribution function of the type $f(x)=(1+kx^2)^{-2}$ (Cauchy square). The dislocation densities derived from the different experiments of the present work by the procedures mentioned above are presented in Table 3. The agreement between the results is very good.

Dislocation densities determined by TEM after deformation at 77 K could not be found in the literature up to now. In Zr–O alloys with dislocation cell structures observed after a strain of 4% at 623 K, Kelly & Smith (1973) obtained total defect densities within the range $(1\text{–}6)\times10^{14}$ m^{-2}, and Holt, Griffith & Gilbert (1987) reported a value of 5×10^{14} m^{-2} for a Zr–25% Nb alloy after cold work (25–30%) at room temperature and subsequent annealing at 670 K. Kohn & Dunne (1977) measured the dislocation densities within subgrains formed by creep at temperatures of 473–873 K ($e=10\%$) and found values of the order of 10^{13} m^{-2}. Similar results were obtained by Dlouhý (1986) for Zr–Sn alloys after deformation at higher temperatures. Although they cannot directly be compared with literature data and an unambiguous control by TEM would be desirable, the values of the dislocation density determined in the present work (Table 3) appear to be reasonable. In this connection it must also be noted that there are several papers which verify the agreement of TEM data and an X-ray analysis of the dislocation density based on the Krivoglaz–Wilkens theory (see references in paper I).

4. Summary and concluding remarks

The present work demonstrates that X-ray diffraction line profile analysis offers a good possibility for a satisfactory quantitative estimation of both the fractions of $\langle a\rangle$ and $\langle a+c\rangle$ dislocations and the dislocation density of hexagonal (or non-cubic structures) even in the case of polycrystalline materials. Moreover, if a number of X-ray diffraction profiles can be measured sufficiently precisely [*e.g.* with the aid of high-resolution X-ray diffractometry (*e.g.* Wilkens & Eckert, 1964; Ungár, 1988)], a more detailed characterization of the dislocation content appears to be possible on the basis of the procedure illustrated by Fig. 8.

This is of particular interest for investigations of samples such as, for instance, technically deformed polycrystalline materials or fine-grained powders, where the application of TEM becomes very difficult or impossible. But since a statistically representative quantitative characterization of the dislocation content even by TEM is not always a simple problem, the X-ray profile analysis should also be taken into account as a suitable complementary tool.

In the practice of the X-ray profile analysis of hexagonal (or other non-cubic) materials it must be taken into consideration that stacking faults and/or twins can be present within the scattering structure, which also give rise to an anisotropy of the diffraction line broadening. In such cases the analysis of the dislocation content should be based on reflections which are unaffected by stacking faults and/or twins, or the dislocation-induced line broadening has to be separated by formal application of the Warren–Averbach analysis.

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