

## AN ANALYTICAL METHOD FOR THE QUANTITATIVE DETERMINATION OF THE VOLUME FRACTIONS IN FIBER TEXTURES

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*Abstract:* In order to evaluate pole-figure measurements quantitatively, one needs the normalization factor which reduces measured intensity values to multiples of the random density. This factor may be determined experimentally by measuring the intensities of a random sample or it may be calculated by integrating over the whole pole-figure or its asymmetric unit. If pole-figure values are not available in the whole angular range  $0 \leq \phi \leq 90^\circ$  (incomplete pole-figures), then the calculation is in general much more difficult and it usually presumes the knowledge of several pole-figures.

In the case of fiber textures (axial symmetry), consisting of only a few strongly preferred orientations with the crystal directions  $\langle uvw \rangle_i$  parallel to the axis of symmetry, the normalization factor and hence the volume fractions of the components  $i$  may be calculated in a rather simple way requiring only one, possibly incomplete, pole figure.

### INTRODUCTION

Fiber textures are orientation distributions of crystallites having axial symmetry. The axis may be the fiber axis in fibers or wires, or the normal direction to the layer plane in evaporated or electrolytically deposited layers.<sup>1</sup> Such textures may occur as strongly preferred orientations, that is, a certain crystal direction  $\langle uvw \rangle$  is oriented nearly parallel (within a few degrees) to the axis of symmetry. In multicomponent fiber textures several different  $\langle uvw \rangle$ -directions

are parallel to the axis of symmetry. Then the problem arises how to determine the relative volume fractions of these components. This problem is easy to solve by integration if the pole-figure is known in the whole angular range  $0 \leq \phi \leq 90^\circ$ . This is, however, often not the case. Pole-figures of electrolytically deposited layers are usually available only in a smaller range  $0 \leq \phi_{\max} < 90^\circ$ . For this case, a general analytical method leading to the inverse pole-figure has been developed on the basis of a series expansion,<sup>2,3</sup> which requires several pole-figures to be measured. If the texture consists of several strongly preferred orientations with separated peaks in the pole-figures then it is possible to calculate the normalization factor and the relative volume fractions in a much simpler way from only one single pole-figure. This method has been used in the case of non-superimposed peaks of the different texture components.<sup>4,5</sup> It can, however be generalized to the case of peaks composed of contributions of several texture components.

#### CALCULATION OF THE NORMALIZATION FACTOR

We fix a spherical polar coordinate system  $\phi\gamma$  within the specimen (Figure 1) in such a way that the direction  $\phi = 0$  is

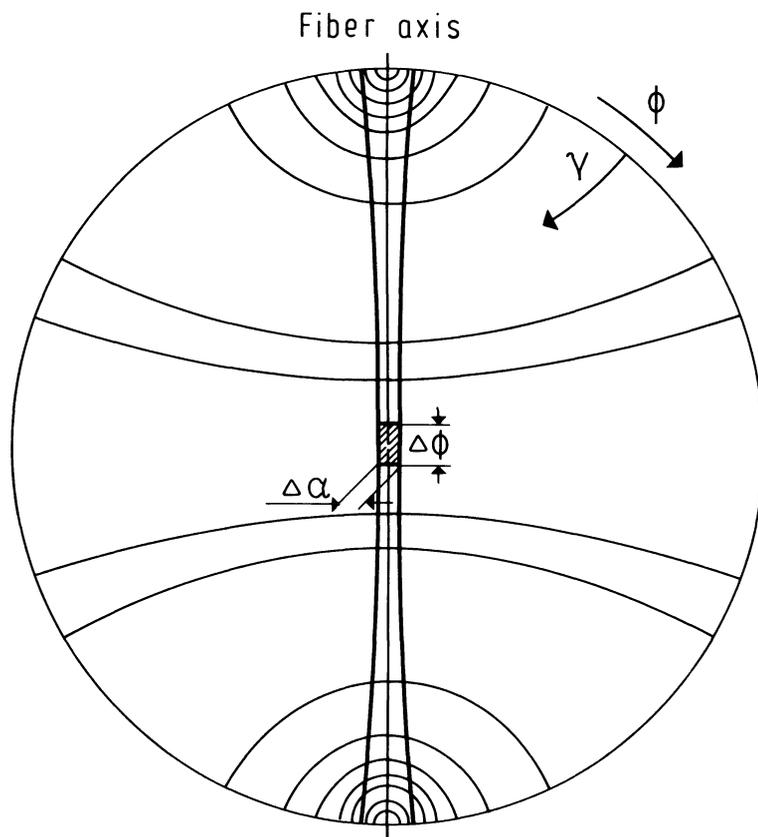


Figure 1. The polar coordinates and the element of solid angle used for the measurement of an axially symmetric texture.

parallel to the fiber axis (axis of symmetry). The orientation distribution of the  $\langle hkl \rangle$ -directions of the crystallites, the  $(hkl)$ -pole-figure is then defined by

$$\frac{dV}{V} = \frac{1}{4\pi} P_{hkl}(\phi\gamma) \sin\phi \, d\phi \, d\gamma \quad (1)$$

where  $dV$  is the volume of the crystallites having their  $\langle hkl \rangle$ -directions parallel to the direction  $\phi\gamma$  within the angular range  $d\phi \, d\gamma$ , and  $V$  is the whole volume of the specimen. The pole distribution function or pole-figure  $P_{hkl}(\phi\gamma)$  is normalized in multiples of the random density. The normalization relation is

$$\int_{\phi=0}^{\pi} \int_{\gamma=0}^{2\pi} P_{hkl}(\phi\gamma) \sin\phi \, d\phi \, d\gamma = 4\pi \quad (2)$$

This corresponds to the value 1 for the random distribution. In the case of fiber textures the pole density function is, by definition, independent of the angle  $\gamma$

$$P_{hkl}(\phi\gamma) = P_{hkl}(\phi) \quad (3)$$

This reduces the normalization relation Eq. (2) to

$$\int_{\phi=0}^{\pi} P_{hkl}(\phi) \sin\phi \, d\phi = 2 \quad (4)$$

If one takes into account the center of symmetry due to Friedel's law, then the integration in Eq. (4) can be limited to  $\pi/2$  and one obtains the normalization relation

$$\int_{\phi=0}^{\pi/2} P_{hkl}(\phi) \sin\phi \, d\phi = 1 \quad (5)$$

The usual way to determine pole-figures is to measure them by an X-ray goniometer. The registered X-ray intensity is proportional to the volume fraction of the crystallites having their  $\langle hkl \rangle$ -directions within a certain angular range  $\Delta\phi \, \Delta\alpha$  determined by the experimental conditions (Figure 1). If the orientation distribution is strongly preferred then the actually measured distribution function may be artificially broadened with respect to the true pole distribution because it is "smeared out" over the angular range  $\Delta\phi \, \Delta\alpha$ . But we are not going to discuss this effect here. Generally the exact values of  $\Delta\phi$  and  $\Delta\alpha$  are not known. Hence, the measured X-ray intensity is proportional to an unknown factor

$$I_{hkl}(\phi) = N_{hkl} \cdot P_{hkl}(\phi) \quad (6)$$

the normalization factor  $N_{hkl}$ , which has to be determined in order to obtain normalized pole-density values.

## RANDOM SAMPLE

If a sample with random orientation distribution is available then the normalization factor simply follows from the relation

$$I_{hkl}(\phi)_r = N_{hkl} \quad (7)$$

## COMPLETE POLE-FIGURES

In the case of complete pole-figures the normalization factor can easily be determined by integration over the whole pole-figure.

$$\int_{\phi=0}^{\pi/2} I_{hkl}(\phi) \sin\phi \, d\phi = N_{hkl} \quad (8)$$

This is a well-known method which has also been used in the case of general, non-axial symmetric textures.<sup>6,7</sup>

## INCOMPLETE SINGLE-COMPONENT POLE-FIGURES

We assume a strongly preferred single component texture in which the  $\langle uvw \rangle$ -directions of all the crystallites are parallel to the axis of symmetry  $\phi = 0$  within a few degrees. Then the measured X-ray intensity distribution function  $I_{hkl}(\phi)$  consists of only a few sharp peaks corresponding to different but crystallographically equivalent  $\langle hkl \rangle$ -directions. These directions are concentrated in narrow angular ranges  $j$  from  $\phi_{j1}$  to  $\phi_{j2}$  on the pole figure (e.g., Figure 2). The range  $j$  must contain the corresponding  $\langle hkl \rangle$ -directions of every crystallite in the sample. Hence a normalization relation similar to Eq. (5) must be valid already for the range  $\phi_{j1}$  to  $\phi_{j2}$

$$\int_{\phi=\phi_{j1}}^{\phi_{j2}} P_{hkl}(\phi) \sin\phi \, d\phi = \frac{Z_j}{Z_{hkl}} \quad (9)$$

where  $Z_j$  is the number of crystallographically equivalent  $\langle hkl \rangle$ -directions falling into the range  $\phi_{j1}$  to  $\phi_{j2}$ , and  $Z_{hkl}$  is the whole number of equivalent  $\langle hkl \rangle$ -directions. With Eq. (6), Eq. (9) leads to the relation

$$S_j = \int_{\phi=\phi_{j1}}^{\phi_{j2}} I_{hkl}(\phi) \sin\phi \, d\phi = \frac{Z_j}{Z_{hkl}} N_{hkl} \quad (10)$$

Since the numbers  $Z_j$  and  $Z_{hkl}$  are known from simple geometrical considerations and  $S_j$  can be determined from the measured intensity distributions the normalization factor  $N_{hkl}$

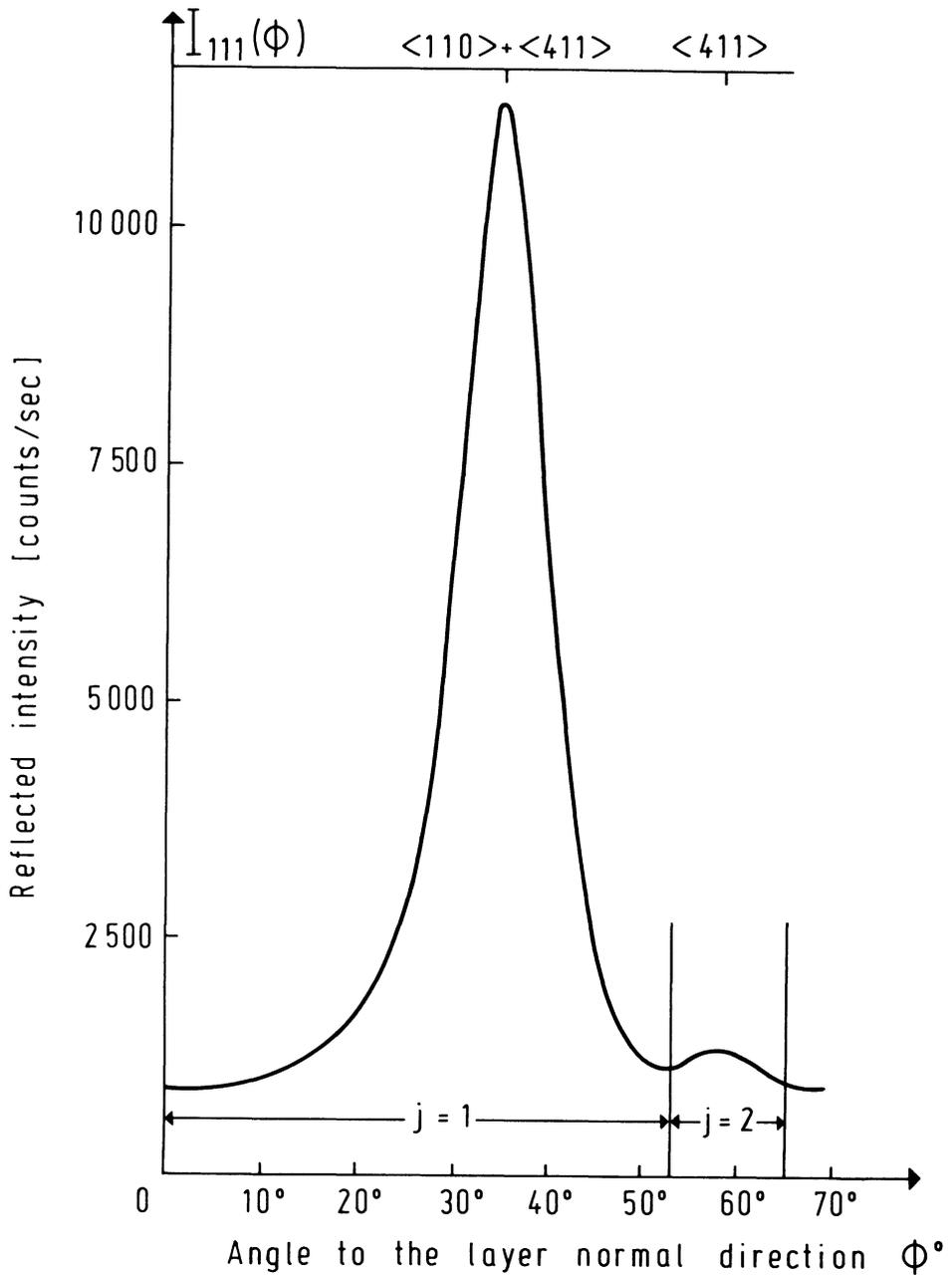


Figure 2. (111)-pole figure of an electrolytically deposited nickel layer (sample 1) according to Table 1.

can be determined by integration over each single peak of the pole-figure.

$$N_{hkl} = Z_{hkl} \frac{S_j}{Z_j} \quad (11)$$

where the ratio  $S_j/Z_j$  does not depend on the number  $j$  of the specific peak.

## INCOMPLETE MULTI-COMPONENT POLE-FIGURES

Now the texture is assumed to be a superposition of several preferred orientations having different crystal directions  $\langle uvw \rangle_i$  parallel to the axis of symmetry. The volume fractions of these components may be  $M^i$  such that

$$\sum_i M^i = 1 \quad (12)$$

We assume at first that the peaks of the different texture components are separate. In this case, one peak of the pole-figure contains the corresponding  $\langle hkl \rangle$ -directions of the crystallites of the  $i$ -component so that the normalization relation Eq. (9) reads

$$\int_{\phi=\phi_{j1}^i}^{\phi_{j2}^i} P_{hkl}(\phi) \sin\phi \, d\phi = \frac{Z_j^i}{Z_{hkl}} M^i \quad (13)$$

where  $Z_j^i$  is the number of equivalent  $\langle hkl \rangle$ -directions of the texture component  $i$  falling into the pole-figure peak number  $j$ . With Eq. (6) one obtains

$$S_j^i = \int_{\phi=\phi_{j1}^i}^{\phi_{j2}^i} I_{hkl}(\phi) \sin\phi \, d\phi = N_{hkl} \frac{Z_j^i}{Z_{hkl}} M^i \quad (14)$$

Eq. (14) gives the following expression for the volume fractions

$$M^i = \frac{Z_{hkl}}{N_{hkl}} \cdot \frac{S_j^i}{Z_j^i} \quad (15)$$

Since the volume fractions  $M^i$  are independent of  $j$ , the number of the specific peak, the ratio  $S_j^i/Z_j^i$  is also independent of  $j$ . Hence the normalization factor can be determined using any one peak of each texture component and summing up over  $i$ , that is over all texture components,

$$N_{hkl} = Z_{hkl} \cdot \sum_i \frac{S_j^i}{Z_j^i} \quad (16)$$

where  $j$  denotes one of the peaks belonging to the texture component  $i$ . Once the normalization factors are known the volume fractions can be obtained from Eq. (15).

## SUPERIMPOSED PEAKS OF DIFFERENT TEXTURE COMPONENTS

If the peak  $j$  contains contributions of two (or more) texture components, say  $i = 1$  and  $i = 2$ , then Eq. (14) is to

be replaced by

$$S_j^{1+2} = \int_{\phi=\phi_{j1}}^{\phi_{j2}} I_{hkl}(\phi) \sin\phi \, d\phi = \frac{N_{hkl}}{Z_{hkl}} [Z_j^1 M^1 + Z_j^2 M^2] \quad (17)$$

which can also be used to determine the volume fractions  $M^i$ , for example, if  $M^1$  is already known from a relation of the type of Eq. (15).

#### APPROXIMATION OF THE INTENSITY DISTRIBUTION BY A LIMITED NUMBER OF MEASURED VALUES

The calculation of the normalization factors and the volume fractions of the texture components is based on the calculation of integrals Eq. (14) over the measured X-ray intensity. Often the intensities are known only at certain equidistant points

$$\phi_m = m \cdot \phi_1 \quad (18)$$

These values may be designated by  $I_m$ . We want to express the Integral  $S$  by the values  $I_m$  only. Since  $\sin\phi$  vanishes at  $\phi = 0$  while  $I_0$  may be a maximum value, the point  $\phi = 0$  has to be treated separately. Hence we put

$$S = \int_{\phi=0}^{\frac{1}{2}\phi_1} I_{hkl}(\phi) \sin\phi \, d\phi + \sum_{m=1}^{m_{\max}} \int_{\phi_m - \frac{1}{2}\phi_1}^{\phi_m + \frac{1}{2}\phi_1} I_{hkl}(\phi) \sin\phi \, d\phi \quad (19)$$

We assume that the step  $\phi_1$  be small compared with the half-maximum width (only in this case can reliable integral values be obtained). Then  $I_{hkl}(\phi)$  may be replaced by  $I_m$  within each interval. In the second integral, also,  $\sin\phi$  is replaced by  $\sin\phi_m$ . In the first integral  $\sin\phi$  is replaced by  $\phi$ . Thus we obtain the following approximation

$$S = \frac{1}{8}(\phi_1)^2 I_0 + \phi_1 \sum_{m=1}^{m_{\max}} \sin\phi_m I_m \quad (20)$$

whereas an integral not including the point  $\phi = 0$  becomes

$$S = \phi_1 \sum_{m_{\min}}^{m_{\max}} \sin\phi_m I_m \quad (21)$$

Eqs. (15), (16) and (17) allow the normalization factors and the volume fractions of the different texture components to be determined with integrals over the intensity values calculated after Eq. (20) or (21).

## EXPERIMENTAL RESULTS

The method was applied to the determination of the amount of growth-twins in electrolytically deposited layers of metals having axially symmetric textures (fiber textures). According to the experimental conditions these layers usually consist of one (or several) relatively sharp matrix orientations  $\langle uvw \rangle$  each of which may contain twins in the orientations  $\langle u'v'w' \rangle_{T_1}$  and may be second order twins  $\langle u''v''w'' \rangle_{T_2}$ . Since the layers are grown on solid substrates, pole figures can only be measured in the back-reflection range  $0 \leq \phi \leq \phi_{\max} < 90^\circ$ . The method described above should therefore be suitable for the determination of the volume fractions of the components. As an example the textures of two nickel samples were studied. The conditions of deposition are given in Table 1. The two

TABLE 1 Textures and experimental conditions of two electrolytically deposited nickel layers.

Electrolyte:	NiSO <sub>4</sub> ·7H <sub>2</sub> O	280g/dm <sup>3</sup>
	H <sub>3</sub> BO <sub>3</sub>	30g/dm <sup>3</sup>
	KCl	20g/dm <sup>3</sup>

Sample	pH-value	Temperature t (°C)	Current density (A/dm <sup>2</sup> )	Texture components $\langle uvw \rangle$
1	4.5	25	5	110 + 411
2	4.5	25	3	110 + 100 + 221

textures consisted of the matrix orientation  $\langle 110 \rangle$  with first order twins  $\langle 411 \rangle_{T_1}$  and of a double matrix orientation  $\langle 110 \rangle + \langle 100 \rangle$  with a first order twin orientation of the second component  $\langle 221 \rangle_{T_1}$  respectively. Pole figures were measured with a Philips texture goniometer using CuK $\alpha$  radiation in 1° steps in the range of  $-70^\circ \leq \phi \leq +70^\circ$ . They were corrected for background scattering. The two (111)-pole figures are shown in Figures 2 and 3, respectively. In both pole figures at least one of the texture components is superimposed by other ones. Hence Eq. (17) dealing with superposed peaks must be employed in addition to Eq. (14) for single component peaks. According to Figure 2, the integrated intensities of the two peaks may be written

$$S_1 = \frac{N_{111}}{Z_{111}} [Z_1^{\langle 110 \rangle} \cdot M^{\langle 110 \rangle} + Z_1^{\langle 411 \rangle} \cdot M^{\langle 411 \rangle}] \quad (22)$$

$$S_2 = \frac{N_{111}}{Z_{111}} \cdot Z_2^{\langle 411 \rangle} \cdot M^{\langle 411 \rangle} \quad (23)$$

where  $Z_{111} = 4$  is the number of all  $\langle 111 \rangle$  poles.  $Z_1^{\langle 110 \rangle} = 2$  is the number of  $\langle 111 \rangle$  poles of the  $\langle 110 \rangle$  component falling

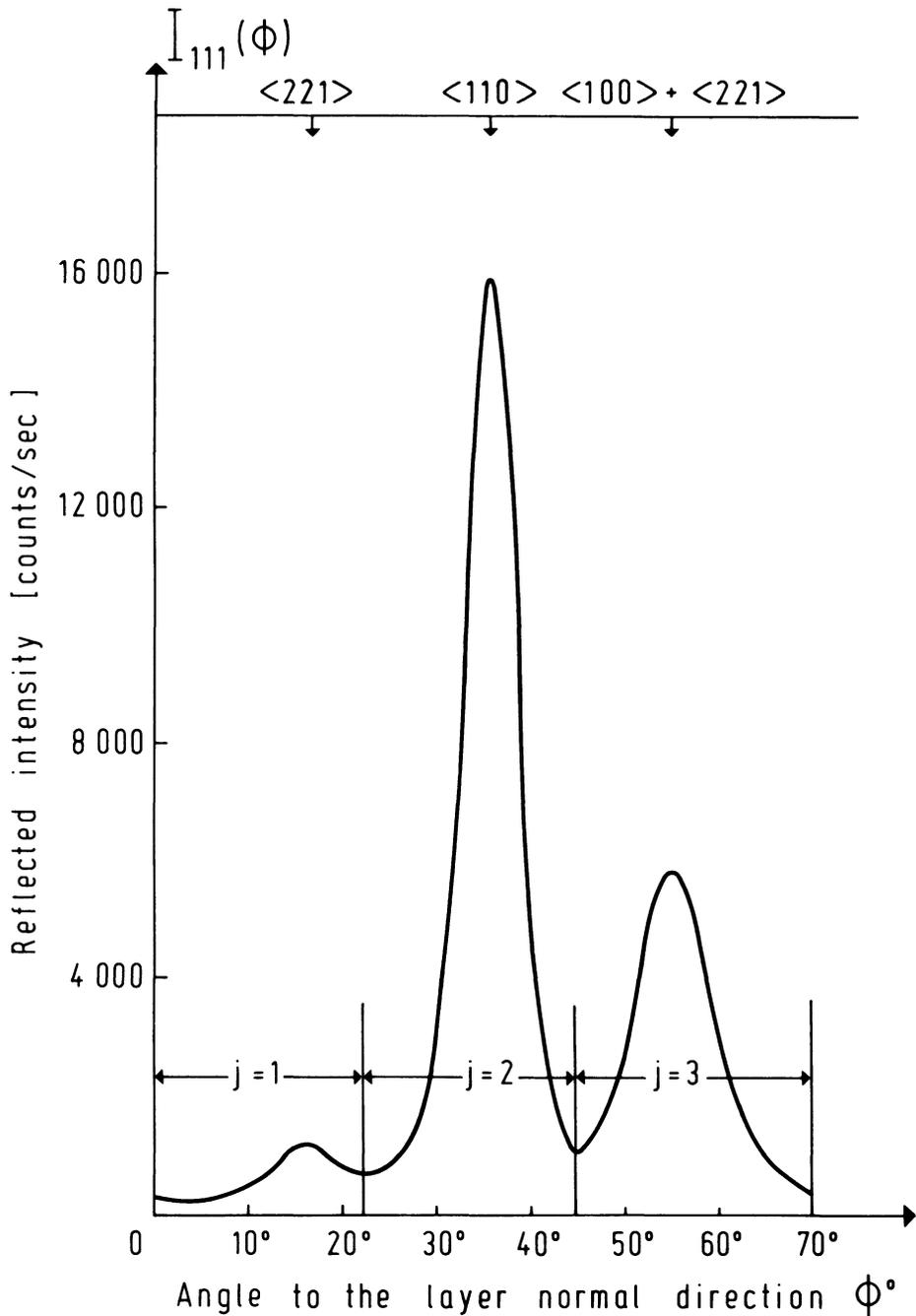


Figure 3. (111)-pole figure of an electrolytically deposited nickel layer (sample 2) according to Table 1.

into the peak  $j = 1$ , and  $Z_1^{<411>} = Z_2^{<411>} = 1$  is the number of  $\langle 111 \rangle$  poles of the  $\langle 411 \rangle$  component falling into each of the two peaks. One thus obtains

$$S_1 = \frac{N_{111}}{4} [2 M^{<110>} + M^{<411>}] \quad (24)$$

$$S_2 = \frac{N_{111}}{4} \cdot M^{<411>} \quad (25)$$

From which it follows

$$S_1 - S_2 = \frac{N_{111}}{4} \cdot 2 M^{<110>} \quad (26)$$

From Eqs. (25) and (26) it follows with Eq. (12)

$$2 S_2 + 2 S_1 = N_{111} [M^{<411>} + M^{<110>}] = N_{111} \quad (27)$$

And for the volume fractions it follows with Eqs. (25) and (26)

$$M^{<411>} = \frac{2 S_2}{S_1 + S_2}, \quad M^{<110>} = \frac{S_1 - S_2}{S_1 + S_2} \quad (28)$$

Similar relations may be easily deduced for the specific situation shown in Figure 3 as well as for the other two pole-figures, the (200) and (220) pole-figures which were also measured. The so obtained volume fractions are given in Table 2. In Table 3 the relative intensities

$$(I_{hkl})_{rel} = \frac{N_{hkl}}{N_{111}} \quad (29)$$

calculated from the so obtained normalization factors are compared with corresponding values obtained experimentally with a random sample, and theoretically according to Sirota.<sup>8</sup> The agreement is quite good.

TABLE 2 Calculated volume fractions of the texture components

Sample	Volume fraction M	Pole-figure			Mean value
		(111)	(200)	(220)	
1	$M^{<110>}$	0.882	0.896	0.912	0.897
	$M^{<411>}$	0.118	0.104	0.088	0.103
2	$M^{<110>}$	0.608	} 0.698	0.664	0.275
	$M^{<221>}$	0.109		0.095	
	$M^{<100>}$	0.283	0.302	0.241	

TABLE 3 Measured and calculated relative intensities.

Pole-figure	Relative intensities			
	Sample 1	Sample 2	Random sample	Calculated
(111)	1.000	1.000	1.000	1.000
(200)	0.429	0.431	0.441	0.450
(220)	0.244	0.234	0.230	0.244

It is a presumption of the method applied here that the peaks belonging to the different texture components are well separated from one another. From Figures 2 and 3 it may be estimated to which degree this presumption is fulfilled. In both cases there is a certain overlapping of peaks which may be one of the reasons for the slight deviations of the obtained values of the volume fractions in Table 2.

The analytical method described here for the determination of the volume fractions of fiber texture components gives reasonable results, and it is much easier to apply than the inverse pole figure method for incomplete pole figures.

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