Fig. 2. Image segmentation and localization by the edge-detection and the edge-coordinate-average methods. (a) The atomic image of the InP phase along the [110] direction; (b) segmentation and localization results.

users from the author by sending a stamped self-addressed label and four 1.44 Mb diskettes (one diskette is enough for only the source code). We will attach an installation note with the mail. A license agreement has to be signed.

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References


A method for complete generation of back-reflection Laue patterns of any single crystal. By CARLOS MARÍN and ERNESTO DIÉGUEZ, Departamento de Física de Materiales, Universidad Autónoma, 28049 Madrid, Spain

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Abstract
Complete numerical simulations of X-ray back-reflection Laue-grams have been carried out. The main factors that affect the intensities of Laue-gram spots have been evaluated. The proposed procedure is general and can be applied to any crystalline material and for any kind of detector and radiation source. The computational requirements are minimal and inexpensive. Excellent agreement between the experimental and the simulated Laue-grams are observed. To demonstrate the potential of these procedures, the case of LiNbO3 is presented here. Exact replication of the Laue-gram for LiNbO3 has been obtained.

X-ray back-reflection Laue-grams are widely used by scientists and technologists to evaluate the monocrystallinity of the synthesized materials and to obtain precisely oriented crystals for various applications (Cullity, 1978). Theoretical simulation of back-reflection Laue-grams have been carried out by several workers (Christiansen & Gerward, 1989; Cornelius, 1981; Fewster, 1984; Hart & Rietman, 1982; Huang, Christensen & Block, 1971; Krahf-Urban, Butz & Preuss, 1973; Laugier & Filhol, 1983; Lisboa & Edwards, 1973; Marín, Cintas & Diéguez, 1994; Ploc, 1978; Preuss, 1979; Reid, 1993; Riquet & Bonnet, 1979), employing different models and algorithms. From these studies, one can obtain the Laue-gram for a specific orientation and the index of any experimental spot. However, a simulation of the experimentally observed Laue-gram with
location and intensity of every spot has not been obtained until now. This hinders its application for the unambiguous indexing of anisotropic crystals and for the automation of the orientation process. An attempt has been made in this direction (Christiansen & Gerward, 1989) but the results obtained are valid only for experimental conditions at selected radiation intensities with some critical wavelengths. For example, when a cathodic source is employed, the wavelengths proposed as critical ones are the characteristic lines of the material used as source. This leads to the reduction in information usually obtained from Laue patterns. The use of these critical intensities does not make use of the fundamental properties of a Laue-gram, which is essentially based on the many-diffraction phenomena produced with reduced exposure time by a continuum of wavelengths.

The model proposed in this paper estimates the relative intensity of each spot of an experimental Laue-gram and can be included as a subroutine in the previously cited programs (Christiansen & Gerward, 1989; Cornelius, 1981; Krahl-Urban, Butz & Preuss, 1973; Laugier & Filhol, 1983; Marin, Cintas & Dieguez, 1994), including the commercial program ORIENTEXPRESS (Cyberstar S.A., 15 Avenue des Martyrs, BP 85X, 38041 Grenoble CEDEX, France). It should be remembered that, since this subroutine must be invoked for each hkl reflection, an optimum computational procedure is under development. The novelty of this procedure is based on a spectral discretization of the continuous radiation. With the errors in computation, the time for calculation and the accuracy of the parameters used from International Tables for X-ray Crystallography (1962) taken into account, an optimal interval of 0.04 Å has been employed for the continuous spectral decomposition.

The following factors and effects have been incorporated in the present model:

1. Spectral sensitivity of the detector; in the case of photographic films, it is necessary to use the zonal distribution of the chemical emulsion.
2. The real spectral intensity of incident radiation on the sample; the source emission, the transmittance and thickness of the filters and of other experimental components must be taken into account.
3. The form factor $f_n$ of ions or atoms, which is a function of the sine function of the incident angle formed by a particular (hkl) plane and the wavelength of incident radiation.
4. The temperature $T$, which modifies the value of $f_n$ according to the expression $f_{nT} = f_n(sin \theta/\lambda) \times exp \left[ -M(sin \theta/\lambda, T, m_n, \Theta_0) \right]$ (Warren, 1990), where $m_n$ is the weight of the atom or ion $n$ and $\Theta_0$ is the Debye temperature of the material; all of them are related by the $M$ function (Warren, 1990).
5. The anomalous dispersion (Cullity, 1978), taking into account the resonance effect of the wavelength which coincides with the absorption lines $K$ of the $n$ atoms of the material. This modifies each form factor to an effective value $f_{n\text{eff}} = f_{nT} + \Delta f(\lambda/\lambda_K)$, where $\Delta f$ is a function of incident wavelength $\lambda$ and $\lambda_K$.
6. The structure factor $F$ (Cullity, 1978), with the following value for the square of its modulus (which is proportional to the emission scattering intensity)

$$|F|^2 = \sum_{n=1}^{N} f_{n\text{eff}} \sin 2\pi(hx_n + ky_n + lz_n) + \sum_{n=1}^{N} f_{n\text{eff}} \cos 2\pi(hx_n + ky_n + lz_n),$$

where $N$ is the number of atoms per unit cell, $h, k, l$ are the indexes of the plane considered, $(x_n, y_n, z_n)$ are the relative coordinates of every atom $n$ of the unit cell and $f_{n\text{eff}}$ is the effective form factor for each element.

7. The absorption factor $AF$, which takes into account the variation of incident and diffracted beams along the sample. This is related to the linear absorption coefficient. If a flat exposure face is used, the expression for $AF$ can be developed by integrating the absorption along the path of the incident ($x$) and the diffracted $[x/\cos(\pi - 2\theta)]$ beam and by considering the geometrical dependence of the diffraction volume $[x \cos(\pi/2 - \theta)]$, so

$$AF \propto \frac{1}{x_{\text{diss}} - x_{\text{inc}}} \int_{x_{\text{inc}}}^{x_{\text{diss}}} \exp \left[ -\mu(x + x/\cos(\pi - 2\theta)) \right] \frac{1}{\cos(\pi/2 - \theta)} dx$$

The coefficient $\mu$ has a spectral dependence, and in a first-order approximation can be calculated (Cullity, 1978) by weighting the coefficients for each of the atoms or ions that constitute the material.

8. The polarization factor $PF$ (Cullity, 1978), due to the geometrical dependence induced in the diffracted intensity by an incident non-polarized radiation. The relative expression is given by $PF = (1 + \cos^2 2\theta)$. This term must be recalculated if we are using polarized radiation.

9. The Lorentz factor $LF$ (Buras & Gerward, 1975), which appears when one takes into account the effect produced by dimensions of collimated beam and distribution of intensities of incident beam around the incident angle. The expression calculated in this geometry is $\lambda^2 \sin 2\theta$. However if we take a spectral interval (which is 0.04 Å in our case), the wavelength range of the diffracted beam must be considered. Thus, the former expression should be multiplied by $\Delta \lambda = \lambda \cot \theta$, and so the Lorentz factor is $LF = \lambda^2 / \sin^2 \theta$ [the same as that of Buras & Gerward (1975) for large perfect crystals].

All the above factors must be considered in a complete model. Moreover, it is necessary to take into account the effective reflection of the planes (hk, nk, nl) because the direction of diffraction of a plane (hkl) contains several orders of diffraction. In this case, following Bragg’s law, $n$ is the order of diffraction considered, with values from 1 to the maximum, which will be given by the minimum wavelength of radiation, the interplanar distance and the incident angle.

The relative intensity of each spot is evaluated by multiplying all these factors. To develop the visual representation of the Laue-gram it is also necessary to know the detector characteristics and to put forward a minimum and a saturation level for the detection using the exposure time as the criterion. In the case of a film detector, the scaling factor between these two levels can be considered linear.

Table 1 summarizes the numerical values employed to perform the simulation. The first block indicates the experimental conditions required for simulating. The parameters of the second block are inherent for a particular material and will be stored in the computer memory. The third block are the data found in International Tables for X-ray Crystallography (1962) and will also be stored permanently in the computer.

Figs. 1 and 2 show the Laue-grs for LiNbO3 crystals. For the experimental Laue-gram, single crystals of LiNbO3 grown...
Table 1. *A priori data required for simulation*

1. Spectral intensity of incident radiation
   - Spectral sensitivity of the detector
   - Orientation to perform
   - Distance from the sample to the centre of the detector

2. Crystalline system of the sample
   - Unit cell parameters and angles
   - Number of atoms of the unit cell (N)
   - Debye temperature

3. Atomic weight of each atom of the material
   - Wavelength of the absorption K edge of each atom
   - Form factor of each kind of atom
   - Linear absorption coefficient per weight of each atom

by the Czochralski method have been used (Martínez-López, Caballero, Santos, Arizmendi & Diéguez, 1993; Santos, Rojo, Arizmendi & Diéguez, 1994). For the calculation, the well known general (International Tables for X-ray Crystallography, 1962) and particular data (Weis & Gaylord, 1985; Abrahams, Reddy & Bernstein, 1966) have been used, with the hexagonal notation and lattice parameters $a = 5.148$ and $c = 13.863 \text{ Å}$ and with a Debye temperature of 503 K.

From the comparison of the Laue-grams, one can conclude that there is an excellent agreement between the simulated and the experimental results, which in turn confirms the rigorous nature of the method developed. The small difference that appears in a few spot intensities could be related to multiple-diffraction phenomena. Some models are under investigation for treatment by the back-reflection Laue technique. Combined with the algorithm previously developed (Marín, Cintas & Diéguez, 1994), this procedure can be used for automatic orientation of any kind of single crystal. Good agreement has been found in other materials like CdTe, Bi$_2$SiO$_5$, Bi$_2$GeO$_2$, Bi$_2$TiO$_3$, Y$_2$SiO$_5$ etc.

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**References**


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Fig. 1. Experimental Laue-gram [sample-to-detector distance 3 cm; plane (2110); molybdenum X-ray tube ($V = 20 \text{kV}, I = 40 \text{mA}$); exposure time 2 h 30 min; film detector Kodak AX].

Fig. 2. Simulated Laue-gram [sample-to-detector distance 3 cm; plane (2110); detection level 0.71% of the Imax; saturation level 6.20% of the Imax; computer 486DX2-66; program language Q-basic; run and storage time 39 s.]
EXTRA: a program for extracting structure-factor amplitudes from powder diffraction data. By A. Altmare, Istituto di Ricerca per lo Sviluppo di Metodologie Cristallografiche, CNR, c/o Dipartimento Geomineralogico, Campus Universitario, Via Orabona 4, 70125 Bari, Italy, M. C. Burla, Dipartimento di Scienze della Terra, Universita\' 06100 Perugia, Italy, G. Cascarano, C. Giaavozzo, A. Guagliardi and A. G. G. Moliterni, Istituto di Ricerca per lo Sviluppo di Metodologie Cristallografiche, CNR, c/o Dipartimento Geomineralogico, Campus Universitario, Via Orabona 4, 70125 Bari, Italy, and G. Polidori, Dipartimento di Scienze della Terra, Universita\', 06100 Perugia, Italy.

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Abstract

Extraction of structure-factor amplitudes from a powder diffraction pattern is not a straightforward procedure. Peak overlapping and background estimation are the main obstacles to the process: they may introduce strong correlations among reflection intensities and heavy errors in their estimates. The program EXTRA is described, which, on the basis of the Le Bail algorithm, is able reliably to estimate the structure-factor amplitudes in a fully automatic way.

1. Introduction

The primary step in any ab initio crystal structure determination process from powder data is the indexing of the reflection peaks and the estimation of the lattice parameters. When this has been done, the next crucial point is the decomposition of the powder diffraction pattern into individual unique integrated intensities. The success of the procedures devoted to the crystal structure solution mainly depends on the accuracy of such a decomposition: poor results should be expected in correspondence with a poor decomposition process. The difficulties mainly arise from three different sources:

(a) the peak overlapping, caused by the collapse of the three-dimensional space on to the one-dimensional diffraction pattern;

(b) the definition of the background;

(c) the preferred orientation of the powder specimen.

Very recently (Altmare, Cascarano, Giaavozzo & Guagliardi, 1994) it has been shown that the preferred orientation may be successfully analysed in the absence of any structural information by the use of usual diffractometer data. We do not discuss in this paper the effects of the preferred orientation on the accuracy of the decomposition process and we focus our attention on points (a) and (b). Such points have been analysed by various authors (Pawley, 1981; Toraya, 1986; David, 1987, 1990; Jansen, Peschar & Schenk, 1992; Estermann, McCusker & Baerlocher, 1992; Bricogne, 1991; Gilmore, Henderson & Bricogne, 1991); as a consequence, a variety of procedures may be found for the accomplishment of a full-pattern decomposition process, all of which are variations of two different prototypes.

1.1. The Pawley (1981) method

This is essentially an iterative process that minimizes the sum of squares of the difference between the observed and the calculated profile. In a standard refinement, the set of parameters that are varied includes, besides the parameters modelling background and peak shape, the integrated intensities themselves. The method suffers from severe problems of ill-conditioning caused by the assumption that all the reflections have independent variable intensities that often refine to negative values. Pawley himself was aware of the problem and introduced slack constraints to the least-squares process to force the intensities to assume allowed values. The problem, however, was not solved. Some notable papers tried to obviate it in the following ways.

(a) Since a wild variation of the intensities mostly occurs when two or more reflections strongly (or completely) overlap, peaks that fulfill this condition are treated as a single peak and the overall intensity is equipartitioned among separate peaks (Jansen, Peschar & Schenk, 1992; but see also Toraya, 1986).

(b) The Bayesian approach is used as a natural probabilistic method to impose the positivity constraint (Sivia & David, 1994). The method is consistent with that of French & Wilson (1978) but is generalized so as to include peak overlapping. The method provides meaningful results even when high overlapping occurs.