X-ray line broadening due to an inhomogeneous dislocation distribution

I. Groma
Department of General Physics, Eötvös University Budapest, Budapest Múzeum krt. 6-8, P.O. Box 323, H-1445 Budapest, Hungary
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A general theory of x-ray line broadening caused by dislocations is presented. It is shown that the leading terms determining the shape of the broadened line profile are independent of the actual dislocation distribution. The intensity distribution can be characterized by average parameters of the dislocation configuration, like the dislocation density, and its fluctuation. For the determination of these parameters a generalized form of Wilson's variance method is proposed. [S0163-1829(98)01513-6]

INTRODUCTION

X-ray line broadening analysis is a frequently applied method for the investigation of dislocation structures in different types of crystalline materials. During the last two decades several successful measurements were carried out on deformed single-crystalline and polycrystalline materials (see, e.g., Ungár *et al.*,[1] Hecker *et al.*,[2] and Klimanek and Kuzel[3]) with this technique. More recent developments are high-resolution x-ray diffraction investigations on the dislocation structure of nanostructured Cu by Ungár *et al.*[4] and on the misfit dislocation density in multilayers by Kaganer *et al.*,[5] opening new perspectives in this field.

The first theory for describing the strain-induced Bragg peak broadening was proposed by Warren and Averbach.[6,7] By Taylor expansion of the Fourier coefficients of the peak they have concluded that the strain-induced broadening scales with the space average of the square of the strain. It was pointed out by Krivoglaz *et al.*[8–10] that if the strain is due to dislocations, the Warren-Averbach method cannot be applied. For a completely random dislocation distribution they worked out an analytical expression. A serious problem with their result is, however, that the Fourier coefficients diverge logarithmically if the crystal size goes to infinity. This problem was first resolved by Wilkens[11–16] by introducing the concept of the restricted random dislocation distribution. For small Fourier coefficients the expression derived by Wilkens has the same form as that obtained by Krivoglaz *et al.*, but the crystal size is replaced by the introduced correlation length. Besides the somewhat artificial foundation of the restricted random dislocation distribution a significant shortcoming of the Wilkens theory is that it predicts symmetrical line profiles in contrast with experimental evidence.[1] Although within the framework of the theory of Wilkens the observed asymmetric broadening can be understood in terms of the quasicomposite model,[1] its disadvantage is that it is based on several *ad hoc* assumptions. Furthermore, as was shown by Gaál[17] a randomly distributed polarized dislocation dipole assembly also causes an asymmetric profile, and so asymmetric broadening can appear under more general conditions than those considered in the quasicomposite model. Recently, Levine and Thomson[18] have proposed an alternative model to overcome the problem of crystal size dependence by the introduction of finite size coherent blocks.

In Refs. 19 and 20 the source of the asymmetry was investigated in terms of the dislocation dipole polarization. Under more general conditions than in the earlier models an asymptotic expression was deduced for the Fourier coefficients. The mathematical foundation of the theory outlined in this paper is similar to the model published earlier. However, it is shown that the result obtained earlier is valid only if the dislocation-dislocation correlation is weak enough. The present study is based only on the analytical properties of the displacement field of straight dislocations, and no assumption is made on the actual form of the dislocation distribution. As a result of this, it can be applied for the inhomogeneous dislocation distribution frequently observed in experimental investigations.

For the determination of the three parameters characterizing the inhomogeneous dislocation distribution an evaluation method is proposed which is a generalization of the variance method first proposed by Wilson[21] (see also Langford[22]).

SCATTERED INTENSITY IN TERMS OF DISLOCATION DENSITY FUNCTIONS

Let us consider a system of N uniform atoms positioned at the points $\vec{R}_i$ $i=1,\ldots,N$. Within the framework of the kinematic scattering theory, the scattered intensity $I(\vec{\kappa})$ has the form

$$I(\vec{\kappa})=C\sum_{j,l=1}^{N}\exp[i\vec{\kappa}(\vec{R}_j-\vec{R}_l)], \tag{1}$$

where $\vec{\kappa}$ is the scattering vector, and C is the atomic scattering factor, which can be taken as constant in the problem under consideration. For a deformed single crystal, after neglecting the second-order terms from Eq. (1) one gets that

$$I(\vec{s})=\frac{C}{V}\int dn^3\int dr^3\exp\{2\pi i\vec{g}[\vec{u}(\vec{r}+\vec{n}/2)-\vec{u}(\vec{r}-\vec{n}/2)]\}\times\exp(i\vec{s}\vec{n}), \tag{2}$$

where $\vec{u}(\vec{r})$ is the displacement field, V is the volume of the crystal, and $\vec{s}=\vec{\kappa}-2\pi\vec{g}$. Here the reciprocal lattice vector $\vec{g}$ was introduced. As can be seen $I(\vec{s})$ is proportional to the Fourier transform of the function

$$A(\vec{n})=\frac{1}{V}\int dr^3\exp\{2\pi i\vec{g}[\vec{u}(\vec{r}+\vec{n}/2)-\vec{u}(\vec{r}-\vec{n}/2)]\}, \quad (3)$$

in which $\vec{n}$ is the Fourier parameter. First its properties are investigated.

In order to obtain the form of $A(\vec{n})$ due to a dislocation network, a system of N parallel straight dislocations is considered with line vectors parallel to the z axis. Denoting the position of the ith dislocation by $\vec{r}_i$ in the xy plane, the total displacement field at the point $\vec{r}$ is

$$\vec{u}(\vec{r})=\sum_{j=1}^{N}\vec{u}_{\rm sing}(\vec{r}-\vec{r}_j), \quad (4)$$

where $\vec{u}_{\rm sing}(\vec{r})$ is the displacement field of a single dislocation. After substituting Eq. (4) into Eq. (3) we get that

$$A(\vec{n})=\frac{1}{F}\int\prod_{j=1}^{N}[1-B(\vec{r}-\vec{r}_j,\vec{n})]dr^2, \quad (5)$$

where F is the cross section of the crystal, and the notation

$$B(\vec{r},\vec{n})=1-\exp\{2\pi i\vec{g}[\vec{u}_{\rm sing}(\vec{r}+\vec{n}/2)-\vec{u}_{\rm sing}(\vec{r}-\vec{n}/2)]\} \quad (6)$$

is introduced. Expression (5) can be transformed into a power series of B:

$$A(\vec{n})=\frac{1}{F}\int dr^2\Bigg[1-\sum_{j=1}^{N}B(\vec{r}-\vec{r}_j,\vec{n})+\frac{1}{2}\sum_{j\neq l}^{N}B(\vec{r}-\vec{r}_j,\vec{n})\times B(\vec{r}-\vec{r}_l,\vec{n})-\frac{1}{6}\sum_{j\neq l\neq m}^{N}B(\vec{r}-\vec{r}_j,\vec{n})B(\vec{r}-\vec{r}_l,\vec{n})\times B(\vec{r}-\vec{r}_m,\vec{n})+\cdots\Bigg]. \quad (7)$$

The above form of $A(\vec{n})$ allows the introduction of different order density functions frequently used in statistical mechanics. Taking the statistical average of several dislocation configurations the sums in Eq. (7) can be replaced by weighted integrals, leading to the formula

$$A(\vec{n})=\frac{1}{F}\int\Bigg[1-\int w^{(1)}(\vec{r}_1)B(\vec{r}-\vec{r}_1,\vec{n})dr_1^2+\frac{1}{2}\int\int w^{(2)}(\vec{r}_1,\vec{r}_2)B(\vec{r}-\vec{r}_1,\vec{n})\times B(\vec{r}-\vec{r}_2,\vec{n})dr_1^2dr_2^2-\frac{1}{6}\int\int\int w^{(3)}(\vec{r}_1,\vec{r}_2,\vec{r}_3)\times B(\vec{r}-\vec{r}_1,\vec{n})B(\vec{r}-\vec{r}_2,\vec{n})B(\vec{r}-\vec{r}_3,\vec{n})dr_1^2dr_2^2dr_3^2+\cdots\Bigg]dr^2, \quad (8)$$

where $w^k(\vec{r}_1,\vec{r}_2,\ldots,\vec{r}_k)$ stands for the kth-order density function. It is important that the different order density functions are not independent from each other, namely,

$$w^{(k-1)}(\vec{r}_1,\vec{r}_2,\ldots,\vec{r}_{k-1})=\frac{1}{N-k}\int w^{(k)}(\vec{r}_1,\vec{r}_2,\ldots,\vec{r}_{k-1},\vec{r}_k)dr_k^2. \quad (9)$$

The expressions obtained above are valid only if all the dislocations have the same Burgers vectors. In a deformed crystal the net Burgers vector is close to zero, and so dislocations with both positive and negative signs have to be allowed for. Equation (8) can be easily generalized for this case:

$$A(\vec{n})=\frac{1}{F}\int dr^2\Bigg\{1-\int dr_1^2[w_+^{(1)}(\vec{r}_1)B(\vec{r}-\vec{r}_1,\vec{n})+w_-^{(1)}(\vec{r}_1)B^*(\vec{r}-\vec{r}_1,\vec{n})]+\frac{1}{2}\int dr_1^2\int dr_2^2[w_{++}^{(2)}(\vec{r}_1,\vec{r}_2)B(\vec{r}-\vec{r}_1,\vec{n})\times B(\vec{r}-\vec{r}_2,\vec{n})+w_{--}^{(2)}(\vec{r}_1,\vec{r}_2)B^*(\vec{r}-\vec{r}_1,\vec{n})B^*(\vec{r}-\vec{r}_2,\vec{n})+w_{+-}^{(2)}(\vec{r}_1,\vec{r}_2)B(\vec{r}-\vec{r}_1,\vec{n})B^*(\vec{r}-\vec{r}_2,\vec{n})+w_{-+}^{(2)}(\vec{r}_1,\vec{r}_2)B^*(\vec{r}-\vec{r}_1,\vec{n})B(\vec{r}-\vec{r}_2,\vec{n})]+\cdots\Bigg\}, \quad (10)$$

where $w_+^{(1)}, w_-^{(1)}, w_{++}^{(2)}(\vec{r}_1,\vec{r}_2), w_{--}^{(2)}(\vec{r}_1,\vec{r}_2)$, and $w_{+-}^{(2)}(\vec{r}_1,\vec{r}_2)$ are the one- and two-particle density functions with positive and negative signs, respectively, and the asterisk denotes the complex conjugate.

TERMS IN $A(\vec{n})$ PROPORTIONAL TO THE FIRST-ORDER DENSITY FUNCTIONS

As a first step the leading term of expression (10),

$$f^0(\vec{r},\vec{n})=\int w_+^{(1)}(\vec{r}_1)B(\vec{r}-\vec{r}_1,\vec{n})dr_1^2, \quad (11)$$

is investigated. By the variable substitution $\vec{r}_1\rightarrow\vec{r}-\vec{r}_1$ one obtains

$$f^0(\vec{r},\vec{n})=\int w_+^{(1)}(\vec{r}-\vec{r}_1)B(\vec{r}_1,\vec{n})dr_1^2. \quad (12)$$

As it was proposed by Warren and Averbach[6] one can try to approximate $f^0(\vec{r},\vec{n})$ with a power series. However, as was first pointed out by Krivoglaz and Ryboshapka[8] at $\vec{n}=0$ the second derivative of $f^0(\vec{r},\vec{n})$,

$$\frac{\partial^2}{\partial n_i \partial n_j} f^0(\vec{r},\vec{n})|_{\vec{n}=0} = -\int w_+^{(1)}(\vec{r}-\vec{r}_1) 4\pi^2 g_l \beta_{li}(\vec{r}_1) g_m \beta_{mj}(\vec{r}_1) dr_1^2, \tag{13}$$

is infinite. In the above expression the distortion tensor

$$\beta_{i,j} = \frac{\partial (\vec{u}_{\rm sing})_i(\vec{r})}{\partial r_j} \tag{14}$$

is introduced. The divergence in expression (13) is due to the fact that the distortion created by a dislocation at the distance $|\vec{r}_1|$ is inversely proportional to $|\vec{r}_1|$, and so the integrand in Eq. (13) has a $1/|\vec{r}_1|^2$-type singularity at the vicinity of $\vec{r}_1 = 0$. (For avoiding divergence at $\vec{r}_1 \to \infty$ it is assumed that $w_+^{(1)}$ goes to zero at infinity. This restriction will be lifted later.)

The calculation method proposed here is based on the realization of the fact that if the coefficient before $B(\vec{r}_1,\vec{n})$ in the integral of expression (12) was zero at $\vec{r}_1 = 0$, the divergence in Eq. (13) would not appear. So if instead of $f^0(\vec{r},\vec{n})$ the expression

$$f^1(\vec{r},\vec{n}) = \int \{w_+^{(1)}(\vec{r}-\vec{r}_1) - w_+^{(1)}(\vec{r}-2\vec{r}_1)\} B(\vec{r}_1,\vec{n}) dr_1^2 \tag{15}$$

is considered, the above singularity in the second derivative of $f^1(\vec{r},\vec{n})$ does not appear. The newly defined $f^1(\vec{r},\vec{n})$ and $f^0(\vec{r},\vec{n})$ are not independent from each other. In order to see their relation let us rewrite expression (15) in the form

$$f^1(\vec{r},\vec{n}) = \int w_+^{(1)}(\vec{r}-\vec{r}_1) B(\vec{r}_1,\vec{n}) dr_1^2 - \int w_+^{(1)}(\vec{r}-2\vec{r}_1) \times B(\vec{r}_1,\vec{n}) dr_1^2. \tag{16}$$

With the variable substitution $2\vec{r}_1 \to \vec{r}_1$ in the second term of Eq. (16) one gets

$$f^1(\vec{r},\vec{n}) = \int w_+^{(1)}(\vec{r}-\vec{r}_1) B(\vec{r}_1,\vec{n}) dr_1^2 - \frac{1}{4}\int w_+^{(1)}(\vec{r}-\vec{r}_1) \times B(\vec{r}_1,2\vec{n}) dr_1^2, \tag{17}$$

where the relation

$$B(\vec{r}_1/2,\vec{n}) = B(\vec{r}_1,2\vec{n}) \tag{18}$$

is applied. Equation (18) is a consequence of the fact that the displacement field of any type of straight dislocation in an elastic medium fulfills the relation

$$\vec{u}_{\rm disl}(\vec{r}_2) - \vec{u}_{\rm disl}(\vec{r}_1) = \vec{u}_{\rm disl}(k\vec{r}_2) - \vec{u}_{\rm disl}(k\vec{r}_1), \tag{19}$$

where k is an arbitrary constant (for verification see the dislocation displacement field in an anisotropic media in Ref. 23). From the comparison of Eqs. (12) and (17) we arrive at

$$f^1(\vec{r},\vec{n}) = f^0(\vec{r},\vec{n}) - \frac{1}{4} f^0(\vec{r},2\vec{n}). \tag{20}$$

Since the first two derivatives of $f^1(\vec{r},\vec{n})$ are finite for small $\vec{n}$'s, it can be approximated by a second-order polynomial:

$$f^1(\vec{r},\vec{n}) = \frac{\partial}{\partial n_i} f^1(\vec{r},\vec{n})\bigg|_{\vec{n}=0} n_i + \frac{1}{2}\frac{\partial^2}{\partial n_i \partial n_j} f^1(\vec{r},\vec{n})\bigg|_{\vec{n}=0} n_i n_j = a_i^{(1)} n_i + a_{i,j}^{(2)} n_i n_j. \tag{21}$$

Equations (20) and (21) lead to the function equation

$$f^0(\vec{r},\vec{n}) - \frac{1}{4} f^0(\vec{r},2\vec{n}) = a_i^{(1)}(\vec{r}) n_i + a_{i,j}^{(2)}(\vec{r}) n_i n_j. \tag{22}$$

Its general solution has the form

$$f^0(\vec{r},\vec{n}) = 2a_i^{(1)}(\vec{r}) n_i - \frac{1}{\ln(2)} a_{i,j}^{(2)}(\vec{r}) n_i n_j \ln(\sqrt{n_l n_l}) + C_{i,j}(\vec{r}) n_i n_j, \tag{23}$$

where the term $C_{i,j}(\vec{r}) n_i n_j$ is the solution of the homogeneous equation

$$f^0(\vec{r},\vec{n}) - \frac{1}{4} f^0(\vec{r},2\vec{n}) = 0. \tag{24}$$

As a consequence of this the coefficient $C_{i,j}(\vec{r})$ is not determined by Eq. (22). In order to obtain its value the expression of $f^0(\vec{r},\vec{n})$ in Eq. (12) has to be analyzed. Since the second derivative of

$$t^0(\vec{r},\vec{n}) = f^0(\vec{r},\vec{n}) - \frac{1}{\ln(2)} a_{i,j}^{(2)}(\vec{r}) n_i n_j \ln(\sqrt{n_l n_l}) \tag{25}$$

is already finite at $\vec{n} = 0$,

$$C_{i,j}(\vec{r}) = \frac{\partial^2}{\partial n_i \partial n_j} t^0(\vec{r},\vec{n})|_{\vec{n}=0}. \tag{26}$$

It is important to note that as has been obtained by Krivoglaz and Ryboshapka[8] $C_{i,j}(\vec{r})$ is proportional to the logarithm of the crystal size. However, as will be shown later the higher-order terms in expression (10) can cancel this logarithmic divergence. In the expression of $f^1(\vec{r},\vec{n})$ in Eq. (21) only the formal values of the coefficients $a_i^{(1)}(\vec{r})$ and $a_{i,j}^{(2)}(\vec{r})$ are given. The actual expression for $a_i^{(1)}(\vec{r})$ can be obtained by calculating the first derivative of $f^0(\vec{r},\vec{n})$:

$$a_j^{(1)}(\vec{r}) = i\pi g_l \int w_+^{(1)}(\vec{r}-\vec{r}_1) \beta_{l,j}(\vec{r}_1) dr_1^2, \tag{27}$$

which can be interpreted as

$$a_j^{(1)}(\vec{r}) = i\pi g_l \beta_{l,j}^{\rm tot\,+}(\vec{r}), \tag{28}$$

where $\beta(\vec{r})_{l,j}^{\rm tot\,+}$ is the total distortion at the point $\vec{r}$ created by all the positive sign dislocations.

For determining the coefficient $a_{i,j}^{(2)}(\vec{r})$ on the basis of Eq. (16) the expression

$$a_{j,k}^{(2)}(\vec{r}) = -\int 2\pi^2 \{w_+^{(1)}(\vec{r}-\vec{r}_1) - w_+^{(1)}(\vec{r}-2\vec{r}_1)\} \times g_l \beta_{l,j}(\vec{r}_1) g_m \beta_{m,k}(\vec{r}_1) dr_1^2 \tag{29}$$

has to be analyzed. Introducing the polar coordinates (r_1,φ) for the variable $\vec{r}_1$, Eq. (29) gets the form

$$a^{(2)}_{j,k}(\vec{r})=\lim_{\epsilon\to 0}-\int_{\epsilon}^{\infty}dr^1\int_0^{2\pi}d\varphi 2\pi^2\{w^{(1)}_{+}(\vec{r}-\vec{r}_1)-w^{(1)}_{+}(\vec{r}-2\vec{r}_1)\}g_l g_m\frac{K_{j,k,l,m}(\varphi)}{r^1}, \tag{30}$$

where $K_{j,k,l,m}(\varphi)$ is a trigonometric polynomial of φ determined by the actual form of the displacement field of the dislocation under consideration. In order to avoid the singularity in the integrand around $r_1=0$ a circular area with radius ϵ is excluded from the integral. By the $2r_1\to r_1$ variable substitution in the second term of the integral (30) one obtains

$$a^{(2)}_{j,k}(\vec{r})=\lim_{\epsilon\to 0}-\int_{\epsilon/2}^{\epsilon}dr^1\int_0^{2\pi}d\varphi 2\pi^2 w^{(1)}_{+}(\vec{r}-\vec{r}_1)\times g_l g_m\frac{K_{j,k,l,m}(\varphi)}{r^1}. \tag{31}$$

Since the integral has to be calculated for an area close to the origin of the coordinate system, $w^{(1)}_{+}(\vec{r}-\vec{r}_1)$ can be approximated by its value at $\vec{r}_1=0$, and so

$$a^{(2)}_{j,k}(\vec{r})=-2\pi^2\ln(2)g_l g_m w^{(1)}_{+}(\vec{r})\int_0^{2\pi}K_{j,k,l,m}(\varphi)d\varphi=-\ln(2)\Phi_{j,k,l,m}w^{(1)}_{+}(\vec{r}). \tag{32}$$

$K_{j,k,l,m}(\varphi)$ is a trigonometric polynomial; therefore the value of $\Phi_{j,k,l,m}$ can always be determined analytically. In most applications $\vec{n}$ is parallel to the diffraction vector $\vec{g}$, and so here only the explicit expression for $\Lambda=\Phi_{j,k,l,m}g_j g_k g_l g_m/|\vec{g}|^4$ in case of isotropic media is given (for more details see Ref. 13):

$$\Lambda=\frac{\pi}{2}C|\vec{g}|^2|\vec{b}|^2\ \sin^2\Psi, \tag{33}$$

in which for screw dislocations

$$C=\ \cos^2\Psi \tag{34}$$

and for edge dislocations

$$C=\ \sin^2\Psi\frac{1}{8(1-\nu)^2}[1-4\nu+8\nu^2+4(1-2\nu)\cos^2\gamma], \tag{35}$$

where ν is the Poisson ratio, Ψ is the angle between $\vec{g}$ and the dislocation line vector $\vec{l}$, and γ is the angle between the vectors $\vec{b}-\vec{l}(\vec{l}\vec{b})$ and $\vec{g}-\vec{l}(\vec{l}\vec{g})$.

The method outlined above can be generalized for the determination of higher-order terms in the expansion on $f^0(\vec{r},\vec{n})$. Without going into the details, if we introduce the function series defined by the recursive formula

$$\nu^k(\vec{r},\vec{r}_1)=\nu^{k-1}(\vec{r},\vec{r}_1)-\frac{1}{2^{k-1}}\nu^{k-1}(\vec{r},2\vec{r}_1),\qquad \nu^0(\vec{r},\vec{r}_1)=w^{(1)}_{+}(\vec{r}-\vec{r}_1), \tag{36}$$

the function

$$f^k(\vec{r},\vec{n})=\int\ \nu^k(\vec{r},\vec{r}_1)B(\vec{r}_1,\vec{n})dr_1^2 \tag{37}$$

can be approximated by a kth-order polynomial of $\vec{n}$ since its first k derivatives are finite at $\vec{n}=0$. After similar steps as applied above one arrives at the following structure for $f^0(\vec{r},\vec{n})$:

$$f^0(\vec{r},\vec{n})=a^{(1)}(\vec{r})n+a^{(2)}n^2\ln(n)+C^{(2)}n^2+\sum_{j=3}^{N}[d^{(j)}(\vec{r})n^j\ln(n)+C^{(j)}(\vec{r})n^j], \tag{38}$$

where for the sake of simplicity only the $n=|\vec{n}|$ dependence of $f^0(\vec{r},\vec{n})$ is indicated. The coefficients $d^{(j)}(\vec{r})$ and $C^{(j)}(\vec{r})$ depend on different order derivatives of $w_+(\vec{r})$. Furthermore, it can be shown that

$$\int\ d^{(j)}(\vec{r})dr^2=0. \tag{39}$$

The contribution of dislocation with negative sign Burgers vector to $A(\vec{n})$ can be given by the same type of expression as Eq. (38).

TERMS IN $A(\vec{n})$ PROPORTIONAL TO THE SECOND-ORDER DENSITY FUNCTIONS

In this section the analytical behavior of the terms proportional to the second-order dislocation distribution functions in Eq. (10) is investigated. Let us introduce the function

$$g(\vec{r},\vec{n}_1,\vec{n}_2)=\int\ dr_1^2\int\ dr_2^2 w^2_{++}(\vec{r}-\vec{r}_1,\vec{r}-\vec{r}_2)\times B(\vec{r}_1,\vec{n}_1)B(\vec{r}_2,\vec{n}_2). \tag{40}$$

It can be seen that the third term in Eq. (10) is equal to $g(\vec{r},\vec{n},\vec{n})$. Since for both variables $\vec{n}_1,\vec{n}_2$ $g(\vec{r},\vec{n}_1,\vec{n}_2)$ has the same structure as $f^0(\vec{r},\vec{n})$, for obtaining a serial expansion of $g(\vec{r},\vec{n}_1,\vec{n}_2)$ a procedure similar to that outlined in the previous section can be applied. Without going into the details of the calculation one gets that the leading terms are (only for $\vec{n}$ parallel to $\vec{g}$)

$$g(\vec{r},\vec{n},\vec{n})=-\kappa(\vec{r})n^2+is^{(2)}(\vec{r})n^3\ \ln(n)+iQ_1(\vec{r})n^3+\Lambda^2 w_{++}(\vec{r},\vec{r})n^4\ \ln^2(n)+Q_2(\vec{r})n^4\ \ln(n)+Q_3(\vec{r})n^4, \tag{41}$$

where

$$s^{(2)}(\vec{r})=\pi g_l\frac{n_m}{n}\Lambda\int\{w^{(2)}_{++}(\vec{r},\vec{r}-\vec{r}_1)+w^{(2)}_{++}(\vec{r}-\vec{r}_1,\vec{r})\}\times\beta_{l,m}(\vec{r}_1)dr_1^2. \tag{42}$$

The coefficients $\kappa(\vec{r})$, $Q_1(\vec{r})$, $Q_2(\vec{r})$, and $Q_3(\vec{r})$ are complicated functions of the two-particle density function. Since they do not have an easily interpretable physical meaning, their actual values are not given here.

SERIES EXPANSION OF $A(\vec{n})$ UP TO $\vec{n}^4$ TERMS

From Eqs. (10), (38), (39), and (42) the leading terms in the series expansion of $A(\vec{n})$ have the form

$$A(\vec{n})=1+\Lambda\langle\rho\rangle n^2\ln(n)-\langle C^{(2)}-\kappa'\rangle n^2+\frac{i}{2}\langle s'^{(2)}\rangle n^3\ln(n)$$
$$+i\langle Q_1'\rangle n^3+\frac{1}{2}\Lambda^2\langle\rho'^{(2)}\rangle n^4\ln^2(n)+\langle Q_2'\rangle n^4\ln(n)$$
$$+\langle Q_3'\rangle n^4, \tag{43}$$

where $\langle\cdots\rangle$ indicates the space average $(1/F)\int\cdots d^2r$,

$$\rho(\vec{r})=w_+^{(1)}(\vec{r})+w_-^{(1)}(\vec{r}) \tag{44}$$

is the dislocation density,

$$\rho^{(2)}(\vec{r})=w_{+,+}^{(2)}(\vec{r},\vec{r})+w_{-,-}^{(2)}(\vec{r},\vec{r})+w_{+,-}^{(2)}(\vec{r},\vec{r})+w_{-,+}^{(2)}(\vec{r},\vec{r}) \tag{45}$$

is the two-particle total dislocation density, and

$$s'^{(2)}(\vec{r})=i\pi g_l\frac{n_m}{|\vec{n}|}\Lambda\int k^{(2)}(\vec{r},\vec{r}-\vec{r}_1)\beta_{l,m}(\vec{r}_1)dr_1^2, \tag{46}$$

in which the notation

$$k^{(2)}(\vec{r}_1,\vec{r}_2)=w_{+,+}^{(2)}(\vec{r}_1,\vec{r}_2)+w_{+,+}^{(2)}(\vec{r}_2,\vec{r}_1)-w_{-,-}^{(2)}(\vec{r}_1,\vec{r}_2)$$
$$-w_{-,-}^{(2)}(\vec{r}_2,\vec{r}_1)+w_{-,+}^{(2)}(\vec{r}_1,\vec{r}_2)-w_{-,+}^{(2)}(\vec{r}_2,\vec{r}_1)$$
$$-w_{+,-}^{(2)}(\vec{r}_1,\vec{r}_2)+w_{+,-}^{(2)}(\vec{r}_2,\vec{r}_1) \tag{47}$$

is introduced. The coefficients $\kappa'(\vec{r})$, $Q_1'(\vec{r})$, $Q_2'(\vec{r})$, and $Q_3'(\vec{r})$ have the same meaning as the corresponding coefficients without a prime in Eq. (42), but they contain the contributions of each type of two-particle density function. It is important to note that the first term in Eq. (38) proportional to n does not have a contribution to $A(\vec{n})$ because the space average of the distortion created by a dislocation is zero. Another important feature of expression (43) is that the coefficient of the term n^2 is the difference of $\langle C^{(2)}\rangle$ and $\langle\kappa'\rangle$. As it was mentioned earlier $\langle C^{(2)}\rangle$ diverges logarithmically with the crystal size. However, if it is assumed that due to the correlation between the dislocations $\langle\kappa'\rangle$ shows the same type of divergence, it can cancel the crystal size dependence which appears without taking into account the higher-order distribution functions. A similar result has been obtained first by Wilkens[13] for the restricted random dislocation distribution and by Gaál[17] for dislocation dipoles. The advantage of the analysis outlined above is that the result is obtained independently from the actual form of the dislocation correlation function.

It is useful to rewrite expression (42) in logarithmic form. By applying the identity:

$$\ln(1+\alpha_1+\alpha_2+\cdots)=\alpha_1+\alpha_2-\frac{1}{2}\alpha_1^2\cdots, \tag{48}$$

we conclude that up to fourth order in n

$$\ln[A(\vec{n})]=\Lambda\langle\rho\rangle n^2\ln\left(\frac{n}{\overline{R_1}}\right)+i\langle s^{(2)}\rangle n^3\ln\left(\frac{n}{\overline{R_2}}\right)+\frac{1}{2}\Lambda^2[\langle\rho^{(2)}\rangle$$
$$-\langle\rho\rangle^2]n^4\ln\left(\frac{n}{\overline{R_3}}\right)\ln\left(\frac{n}{\overline{R_4}}\right), \tag{49}$$

where $\overline{R_1}$, $\overline{R_2}$, $\overline{R_3}$, and $\overline{R_4}$ are parameters with length dimension. Their actual values are complicated functions of the different order dislocation density functions. The advantage of the logarithmic form is that if the dislocation distribution is nearly random, the value of $\langle\rho^{(2)}\rangle-\langle\rho\rangle^2$ is smaller than $\langle\rho^{(2)}\rangle$ and so in Eq. (49) the term proportional to $n^4\ln(n/\overline{R_3})\ln(n/\overline{R_4})$ can be neglected beside $n^2\ln(n/\overline{R_1})$ up to larger n values than in Eq. (43).

The generalization of the above result for more than one type of dislocation system is straightforward. It can be obtained that in Eq. (49) Λ has to be replaced by the average of the Λ values of the different dislocations.

WEAK CORRELATION

It is useful to investigate the values of the parameters in Eq. (49) if the correlations are weak in the dislocation system. A weak correlation means that the contributions of correlation functions

$$T_{i,j}(\vec{r}_1,\vec{r}_2)=w_{i,j}^{(2)}(\vec{r}_1,\vec{r}_2)-w_i^{(1)}(\vec{r}_1)w_j^{(1)}(\vec{r}_2),$$
$$i,j=+,-, \tag{50}$$

to the values of $\langle\rho^{(2)}\rangle$ and $\langle s^{(2)}\rangle$ are small; i.e., from this point of view the two-particle distribution functions can be approximated by the product of two one-particle distribution functions. The correlations, however, can never be completely negligible, because it would result in a crystal size dependence as was discussed earlier.

If $T_{i,j}(\vec{r}_1,\vec{r}_2)$ is neglected in Eqs. (44) and (45), we find that

$$\rho^{(2)}(\vec{r}_1,\vec{r}_2)=\rho(\vec{r}_1)\rho(\vec{r}_2), \tag{51}$$

and so

$$\langle\rho^{(2)}\rangle-\langle\rho\rangle^2=\langle(\rho)^2\rangle-\langle\rho\rangle^2=\langle(\rho-\langle\rho\rangle)^2\rangle. \tag{52}$$

This means that in the case of a weak correlation in Eq. (49) the coefficient of the term $n^4\ln^2(n)$ is proportional to the scatter of the dislocation distribution. In a similar way for a weak correlation from Eqs. (46) and (47),

$$\langle s'^{(2)}\rangle=2\Lambda\pi g_l\frac{n_m}{n}\int dr^2\int dr_1^2\rho(\vec{r})\{w_+(\vec{r}-\vec{r}_1)$$
$$-w_-(\vec{r}-\vec{r}_1)\}\beta_{l,m}(\vec{r}_1)$$
$$=2\Lambda\pi g_l\frac{n_m}{n}\langle\beta^{\rm tot}\rho\rangle. \tag{53}$$

BEHAVIOR OF THE DIFFERENT ORDER MOMENTS OF THE INTENSITY DISTRIBUTION

In the previous sections the analytical behavior of the Fourier transform of the Bragg peak was investigated. In this section the connection between the obtained analytical expression (43) and the different order moments of the scattered intensity distribution $I(\vec{s})$ is established. As will be demonstrated later this leads to an evaluation method which makes it possible to determine the characteristic parameters of the dislocation distribution with less scatter than the earlier applied ones.

Since in most investigations only the dependence of the complete $I(\vec{s})$ on the diffraction angle θ is determined, in the following only the reduced intensity distribution

$$I(q)=\int I(q\vec{g}/|g|+\vec{s}_{\perp})d^2s_{\perp} \tag{54}$$

will be considered. In the above expression the integration has to be carried out for a plane perpendicular to the diffraction vector $\vec{g}$, and $q=4\pi/\lambda[\sin(\theta)-\sin(\theta_0)]$ where θ_0 is the Bragg angle.

As is known the kth-order moment of the scattered intensity

$$m_k=\int_{-\infty}^{\infty}q^kI(q)dq \Big/ \int_{-\infty}^{\infty}I(q)dq \tag{55}$$

can also be determined from the Fourier transform of $I(q)$. Namely,

$$m_k=(i)^k\frac{1}{A(0)}\frac{d^k}{dn^k}A(n)\bigg|_{n=0}, \tag{56}$$

which can be proved as follows:

$$\begin{aligned}\int_{-\infty}^{\infty}q^kI(q)dq&=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}q^kA(n)\exp(inq)dndq\\&=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}A(n)(-i)^k\frac{d^k\exp(inq)}{dn^k}dndq\\&=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}(i)^k\frac{d^kA(n)}{dn^k}\exp(inq)dndq\\&=2\pi(i)^k\int_{-\infty}^{\infty}\frac{d^kA(n)}{dn^k}\delta(n)dn\\&=2\pi(i)^k\frac{d^k}{dn^k}A(n)\bigg|_{n=0}.\end{aligned} \tag{57}$$

Since the second and higher derivatives of $A(n)$ given by the expression (43) have infinite values at $n=0$, in case of line broadening due to dislocations the second- and higher-order moments of the scattered intensity are also infinite. As a result of this the moments cannot be directly used for the determination of the characteristic parameters of the dislocation configuration. In order to obtain applicable results the behavior of the quantities

$$v_k(q')=\int_{-q'}^{q'}q^kI(q)dq \Big/ \int_{-\infty}^{\infty}I(q)dq \tag{58}$$

has to be investigated. [$v_k(q')$ is referred to as the kth-order variance.] As a first step the properties of $v_2(q')$ are analyzed. For overcoming the singularities mentioned above let us introduce the function

$$A_1(n)=A(n)-\frac{1}{2^2}A(2n) \tag{59}$$

and its Fourier transform

$$I_1(q)=\int_{-\infty}^{\infty}A_1(n)\exp(inq)dn. \tag{60}$$

The second derivative of $A_1(n)$ at $n=0$ is already finite. Applying the corresponding Eq. (56) type formula and using the expression (43) we obtain that

$$\begin{aligned}\lim_{q'\to\infty}\int_{-q'}^{q'}q^2I_1(q)dq&=-2\pi\frac{d^2}{dn^2}A_1(n)\bigg|_{n=0}\\&=4\pi\Lambda\langle\rho\rangle\ln(2).\end{aligned} \tag{61}$$

On the other hand, from the definition (60) of $I_1(q)$ one can find that

$$\begin{aligned}\int_{-q'}^{q'}q^2I_1(q)dq=\int_{-q'}^{q'}\int_{-\infty}^{\infty}q^2\left(A(n)-\frac{1}{4}A(2n)\right)\\\times\exp(inq)dndq.\end{aligned} \tag{62}$$

Splitting the integral on the right-hand side of the above equation into two integrals and introducing a $n\to 2n$ variable substitution in the second one we get

$$\begin{aligned}\int_{-q'}^{q'}q^2I_1(q)dq&=\int_{-q'}^{q'}q^2\left[I(q)-\frac{1}{8}I\left(\frac{q}{2}\right)\right]dq\\&=\int_{-q'}^{q'}q^2I(q)dq-\int_{-q'/2}^{q'/2}q^2I(q)dq\\&=[v_2(q')-v_2(q'/2)]\int_{-\infty}^{\infty}I(q)dq.\end{aligned} \tag{63}$$

From Eqs. (61) and (63) one can conclude that for q' values large enough the quantity $v_2(q')$ fulfills the equation

$$v_2(q')-v_2(q'/2)=2\Lambda\langle\rho\rangle\ln(2). \tag{64}$$

Its general solution has the form

$$v_2(q')=2\Lambda\langle\rho\rangle\ln\left(\frac{q'}{q_0}\right), \tag{65}$$

where q_0 is a constant the value of which is not determined by Eq. (64). This means that the second-order moment of the line profile obtained on a dislocated crystal diverges logarithmically with q'. Consequently the asymptotic behavior of the intensity distribution has the form

$$I(q)=\Lambda\langle\rho\rangle\frac{1}{|q|^3}+\cdots. \tag{66}$$

A similar result was obtained for a single screw dislocation by Wilson[21] and for a restricted random dislocation distribution by Wilkens.[14] The present analysis shows, however, that the Eq. (66) type decay of the scattered intensity is independent of the actual form of the dislocation distribution.

The asymptotic properties of $v_3(q')$ can be obtained by a similar method outlined above. The only difference is that in this case the behavior of the function

$$A_2(n)=A_1(n)-\frac{1}{2^3}A_1(2n) \tag{67}$$

has to be analyzed in which the function $A_1(n)$ is defined by Eq. (59). It leads to the result that for large q' values

$$v_3(q')=-6\langle s'^{(2)}\rangle\ln\left(\frac{q'}{q_1}\right), \tag{68}$$

where q_1 is a constant the value of which cannot be determined from Eq. (68). It follows that beside the term (66) there is a $q/|q|^5$ antisymmetric term in the decay of the intensity distribution, i.e.,

$$I(q)=\Lambda\langle\rho\rangle\frac{1}{|q|^3}-3\langle s'^{(2)}\rangle\frac{q}{|q|^5}+\cdots\ . \tag{69}$$

In order to determine the contribution of the fourth-order term in $A(n)$ to the intensity distribution the function

$$A_3(n)=A_2(n)-\frac{1}{2^4}A_2(2n)-\frac{1}{2^4}\left(A_2(2n)-\frac{1}{2^8}A_2(4n)\right) \tag{70}$$

and its Fourier transform $I_3(q)$ have to be introduced. Since the first four derivatives of $A_3(n)$ are finite at $n=0$, the fourth-order moment of $I_3(q)$ can be determined from the relation (56):

$$m_4^{(3)}(q')=\int_{-q'}^{q'}q^4I_3(q)dq\Big/\int_{-\infty}^{\infty}I(q)dq=96\Lambda^2\langle\rho'^{(2)}\rangle. \tag{71}$$

which is certainly valid only for large q' values. On the other hand, $m_4^{(3)}(q)$ can be given with $v_4(q)$ as follows:

$$m_4^{(3)}(q')=v_4(q)-8v_4(q/2)+21v_4(q/4)-22v_4(q/8)+8v_4(q/16). \tag{72}$$

For large q values Eqs. (71) and (72) define a function equation for $v_4(q)$, the general solution of which is

$$v_4(q)=c_1q+c_2q^2+12\Lambda^2\langle\rho'^{(2)}\rangle\ln(q/q_2)\ln(q/q_3), \tag{73}$$

where c_1, c_2, q_2, and q_3 are parameters which cannot be determined from Eqs. (71) and (72). However, from Eq. (69) one can find that $c_1=0$ and $c_2=\Lambda\langle\rho\rangle$, i.e., for large enough q values,

$$v_4(q)=\Lambda\langle\rho\rangle q^2+12\Lambda^2\langle\rho'^{(2)}\rangle\ln(q/q_2)\ln(q/q_3). \tag{74}$$

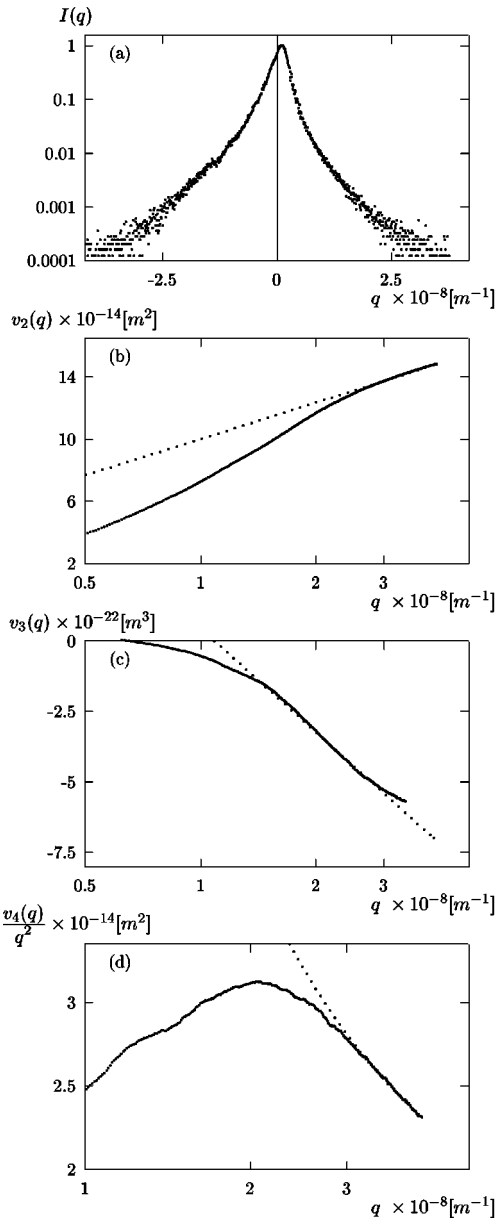

FIG. 1. (a) Line profile measured on a tensile deformed Cu single crystal, $\tau=37$ MPa (Ref. 20). (b) Second-order variance vs q and the fitted expression (65). (c) Third-order variance vs q and the fitted expression (68). (d) Fourth-order variance vs q and the fitted expression (74).

EVALUATION PROCEDURE

On the basis of the theoretical results obtained above the line profile analysis of the measured intensity distribution can be carried out either on the measured profile itself or on its Fourier transform. The fitting of the expression (49) to the Fourier transform of the profile, however, leads often to large scatters in the parameters. This is due to the fact that expression (49) contains seven free parameters and it is valid only for a few Fourier coefficients. Better results can be obtained by the evaluation of the different order variances.

The method proposed here is based on the asymptotic expressions (65), (68), and (74). By fitting the corresponding

expressions to the different order variances of the measured profile for large q values one can determine the dislocation density, the average dislocation density fluctuation, and $\langle s'^{(2)} \rangle$. The method is demonstrated on a line profile [see Fig. 1(a)] measured on a Cu single crystal which was tensile deformed in the [100] direction up to 37 MPa stress. The diffraction vector $\vec{g}=(200)$. (The profile has been published earlier by Ungár *et al.*[20]) Figures 1(b), 1(c), and 1(d) show the second-, third-, and fourth-order variances versus q with the fitted functions (65), (68), and (74) (dotted lines), respectively. It can be seen that for about 100 data points the measured data are very accurately described by the asymptotic formulas. Consequently the scatter in the parameters is significantly smaller compared to the determination through the Fourier coefficients. Another advantage of this method is that it contains the possibility of an internal checking; namely, the dislocation density can be determined from both the second- and fourth-order variances.

CONCLUSIONS

The theory developed in the present work gives an asymptotic analytical expression for the Fourier transform of the broadened x-ray diffraction peak of crystalline materials containing dislocations. It is shown that for a small Fourier parameter the line profile can be characterized by three parameters: the average dislocation density, the average dislocation density fluctuation, and the average of the product of the dislocation density and the total distortion created by the dislocations $\langle \beta^{\mathrm{tot}} \rho \rangle$. Since the third parameter appears in the leading term in the imaginary part of the Fourier transform of the profile, it accounts for the asymmetric broadening and through the distortion it also reflects the internal stresses developed in the crystal. On the basis of the result obtained for the Fourier transform of the broadened profile the asymptotic properties of the scattered intensity are also determined.

The theory outlined gives only the first three terms in the series expansion of the Fourier transform of the Bragg peak and the behavior of the scattered intensity for large q values. Consequently, in contrast to the theories of Krivoglaz *et al.* and Wilkens developed for certain specific dislocation distributions, the entire measured profile cannot be recovered on its basis. A significant advantage of the present description method is that its results are not restricted by any assumption on the form of the dislocation distribution. The obtained results follow directly from the expansion invariance of the displacement fields of straight dislocations expressed by Eq. (19).

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