Asymmetric X-ray line broadening caused by dislocation polarization induced by external load

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Abstract

An external load, even if smaller than the flow stress, generates dislocation rearrangement leading to a net polarization of the system. According to the theory of X-ray line broadening, this polarization leads to Bragg peak asymmetry. Here we present an experimental validation of the theory by in situ profile measurements on “elastically” deformed Cu single crystals. The profiles are evaluated by the “restricted moments method”. In agreement with the theory it is found that peak asymmetry is increasing with deformation.

Key words: X-ray diffraction, plastic deformation, dislocation theory, statistical mechanics, dislocation polarization

The dislocation system developing during plastic deformation of crystalline materials represent a complex network of connected interacting lines, with collective properties that can only be described by statistical physics methods. A few examples, that are in the focus of recent investigations, are dislocation pattern formation, size effects, or dislocation avalanches. The physical description of each of them requires statistical approach.

Beside the mean values of parameters like the dislocation density or the Nye’s tensor, a key quantity for a statistical description of the properties of the dislocation networks is the dislocation-dislocation correlation function [1]. So the experimental determination of such parameters that are directly linked to the correlation function are essential for the better understanding of the collective properties of the dislocations.

Since in most cases the total dislocation density is much larger than the geometrically necessary one, large portion of the dislocations form closed loops which can be characterized by their dipole moments (see [2,3]). On the other hand, however, the dipole moment per unit volume (polarization) is obviously determined by the dislocation-dislocation correlation function. So, dislocation polarization is one of the measure of the correlation properties of a dislocation system.

In this letter first we demonstrate with a simple example how dislocation polarization enters into the field theory of dislocations, then X-ray line profile measurements are presented to determine the polarization properties of deformed Cu single crystals.

As it is well-known, if one considers a dislocation dipole formed by two edge dislocations with opposite signs, in the absence of external load the two dislocations are in equilibrium at 45° angle relative to the slip direction. Applying an external load modifies the relative angle in a reversible manner since the angle is uniquely determined by the load providing that the load level is always smaller than a critical value. (Correspondingly, for loops the external load modifies the dislocation curvature.)

This elementary process plays an important role in the collective properties of dislocation systems. In order to explain the role of the polarization phenomenon let us consider a set of parallel edge dislocations with possible Burgers vectors $(s_i b, 0, 0)$, where $s_i = \pm 1$ is the sign of the ith dislocation. The stress field generated by the dislocation system can be found by the solution of the equation [4]

$$\triangle^2 \chi = \frac{b\mu}{1-\nu} \partial_y \left[\sum_{i=1}^{N} s_i \delta(\boldsymbol{r} - \boldsymbol{r}_i) \right], \tag{1}$$

where $\boldsymbol{r}_i$ is the position of the ith dislocation, μ is the shear modulus, ν is the Poisson’s ratio, and δ is the Delta function. From the stress potential χ the shear stress is $\tau = \partial_x \partial_y \chi$. After introducing a coarse graining length scale (in the order of dislocation spacing) the discrete dislocation

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density

$$\alpha_{\text{discrate}}(\boldsymbol{r}) = \sum_{i=1}^{N} s_i \delta(\boldsymbol{r} - \boldsymbol{r}_i) \qquad (2)$$

can be approximated by a multipole expansion. Up to the first two leading terms

$$\alpha_{\text{discrate}}(\boldsymbol{r}) \approx \alpha(\boldsymbol{r}) + \partial_x P(\boldsymbol{r}), \qquad (3)$$

where $\alpha(\boldsymbol{r})$ is the local Geometrically Necessary Dislocation (GND) density, and $P(\boldsymbol{r})$ is the dislocation polarization [2]. Since according to Fig. 1 there are four possible equivalent dipole configurations, the external stress can generate only net polarization parallel to the slip direction. Assuming

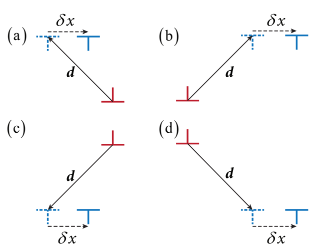

Fig. 1. Four possible dipole configurations (a-d) under external shear.

that the polarization is a unique function of the shear stress Eq. (1) reads as

$$\triangle^2 \chi = \frac{b\mu}{1-\nu} \partial_y \left\{ \alpha(\boldsymbol{r}) + \partial_x P[\partial_x \partial_y \chi(\boldsymbol{r})] \right\}. \qquad (4)$$

It follows, if excess dislocations are present or traction is applied at the surface of the body, due to the induced dislocation polarization the stress function χ fulfills an effective equation. In analogy with dielectrics, for small enough shear stress the $P(\tau)$ relation can be approximated by a linear function, *i.e.*

$$P(\tau) = \lambda \frac{\tau}{b\mu}, \qquad (5)$$

where the parameter λ can be called as the susceptibility of the dislocation system. It should be mentioned that the argument explained above can be extended for 3D dislocation loops in a relatively straightforward manner [2].

As it is demonstrated above, the dislocation polarization induced by the average stress is an important quantity for developing the statistical continuum theory of dislocations that is a hot topic in dislocation theory [5–7]. The experimental determination of the polarization properties of dislocation systems is crucial for the validation of the theory.

X-ray line profile analysis is a powerful method to determine several different statistical parameters of the dislocation network (like total dislocation density, dislocation density fluctuation, internal stress, etc.) [8–11]. As it was first suggested by Gaál [12], dislocation polarization generates asymmetric X-ray line broadening. So, line profile analysis represents a unique experimental method to measure dislocation polarization.

According to the asymptotic theory of line broadening elaborated by Groma *et al.* [9,14], up to the two leading terms the asymptotic part of the intensity distribution $I(q)$ (normalized as $\int I(q)\mathrm{d}q = 1$) reads as

$$I(q) = \frac{\Lambda}{4\pi^2} \frac{\langle \rho \rangle}{|q^3|} + \frac{3}{8\pi^3} \langle s \rangle \frac{q}{|q^5|}, \quad |q| \to \infty, \qquad (6)$$

where $\langle \rho \rangle$ is the average dislocation density, Λ is a constant (in the order of 1) depending on the dislocation and the measuring geometry (for details see [9]), $\langle s \rangle$ is a parameter determined by the dislocation-dislocation correlation as

$$\langle s \rangle = \pi \Lambda \sum_{i,j}^{3} \frac{g_i g_j}{|\boldsymbol{g}|} \int \epsilon_{i,j}(\boldsymbol{r}) T_{+-}(\boldsymbol{r}) \mathrm{d}r^2, \qquad (7)$$

where $\boldsymbol{g}$ is the diffraction vector, $\epsilon_{ij}(\boldsymbol{r}) \propto 1/r$ is the elastic deformation field of a single dislocation, and $T_{+-}(\boldsymbol{r})$ is the correlation function between two dislocations with opposite signs [9]. (For the relation between $\langle s \rangle$ and the dislocation polarization see below.) The diffraction parameter q is defined as $q = 2[\sin(\Theta) - \sin(\Theta_0)]/\lambda_{\text{w}}$, where λ_{w} is the X-ray wavelength, Θ is the diffraction angle, and Θ_0 is the Bragg angle selected for the measurement. It should be mentioned that in the experimental setup used in the measurements the 3D diffraction intensity distribution is integrated for the plane perpendicular to the diffraction vector of the Bragg reflection selected [9]. (This measuring configuration is commonly called "deformation broadening".) It is interesting to note at this point that the internal stress distribution generated by a dislocation system has a similar asymptotic form [16] (for details see below).

Typical intensity distribution measured on a compressed Cu single crystal is shown in Fig. 2. As it is seen, the

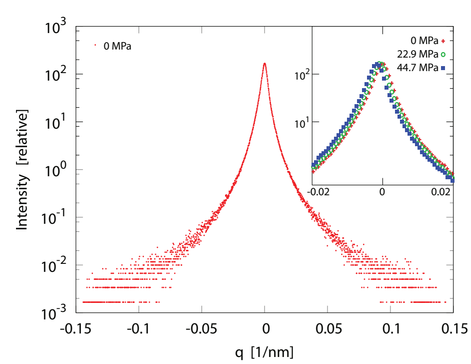

Fig. 2. (200) intensity distribution, obtained on a middle oriented Cu single crystal pre-compressed up to 58.2 MPa. The inset shows the shifting of the peak due to subsequent in-situ external loads of 0, 22.9, 44.7 MPa.

counts at the tail of the intensity distribution have a relatively large scatter, so the direct fitting of the form given by Eq. (6) would result in a large error in the parameters obtained. The error can be reduced considerably if one analyzes the kth order restricted moments [9] defined as

$$v_k(q) := \int_{-q}^{q} q'^k I(q') \mathrm{d}q', \qquad (8)$$

in which q is measured from the center of gravity of the intensity distribution. One can find from Eq. (6) that for large enough q values

$$v_3(q) = \frac{3}{4\pi^3}\langle s\rangle \ln(q/q_1), \tag{9}$$

where q_1 is a constant and

$$f(q) := v_4(q)/q^2 = \frac{\Lambda}{4\pi^2}\langle \rho\rangle. \tag{10}$$

So $f(q)$ tends to a constant proportional to the average dislocation density, while $v_3(q)$ becomes logarithmic in q with a slope proportional to $\langle s\rangle$. This two key features will be used in the data analysis below.

As it was already mentioned, the quantity $\langle s\rangle$ determining the asymptotic asymmetry of the Bragg peak is a quite complicated function of the dislocation-dislocation correlation function [9,14]. It is intuitive, however, to consider a homogeneous polarized dislocation system (Fig. 1). For this simplified case one can find that

$$\langle s\rangle = \pi\Lambda\langle\rho\rangle \sum_{i,j}^{3} \frac{g_i g_j}{|\boldsymbol{g}|}\epsilon_{i,j}(\boldsymbol{d}), \tag{11}$$

where $\boldsymbol{d}$ is the relative position vector of the two dislocations of the dipole [14]. In the absence of the external load the four equally probable possible dipole configurations cancel $\langle s\rangle$. If, however, load is applied one gets a net $\langle s\rangle$ value. It is easy to see that if the load is small enough the mean value of $\langle s\rangle$ is

$$\langle s\rangle = K\langle\rho\rangle\frac{1}{d^2}\delta x, \tag{12}$$

in which K is a constant depending on the dislocation geometry, and δx is the dislocation displacement (see Fig. 1). After introducing the dipole polarization density

$$P = \frac{\langle\rho\rangle}{2}\delta x \tag{13}$$

the above expression gets the form

$$\langle s\rangle = B\langle\rho\rangle P, \tag{14}$$

where $B = K\langle\rho\rangle/d^2$ is a dimensionless constant determined by the dipole distance distribution and the measuring geometry. In the general case, if external load is applied $\langle s\rangle$ changes because of the modification of dislocation correlation function. So, $\langle s\rangle$ is a measure of the response of the system. It should be mentioned at this point, that although the theoretical description of the X-ray line broadening is developed strictly only for straight dislocations, due to the fact that the average dislocation radius of curvature is often much larger than the dislocation spacing, this approximation is practically always justified.

Summarizing the theoretical considerations described above, if one applies an in-situ external load during the line profile measurement, due to the induced dipole polarization the asymmetry of the peak modifies. However, two remarks, important for the further considerations, should be made at this point:

– Long range internal stress that may develop in the crystal during the plastic deformation results in an asymmetric X-ray peak even in the absence of external load [8]. Yet, the level of asymmetry changes if one applies an external load.
– As it is well-known, due to the lattice parameter change caused by the elastic deformation of the crystal, the center of gravity of the intensity distribution shifts when an external load is applied (see the inset of Fig. 2). This is, however, an effect independent from the dislocation content of the crystal. (The shift was determined in the measurements performed for validating the correctness of the measuring setup.)

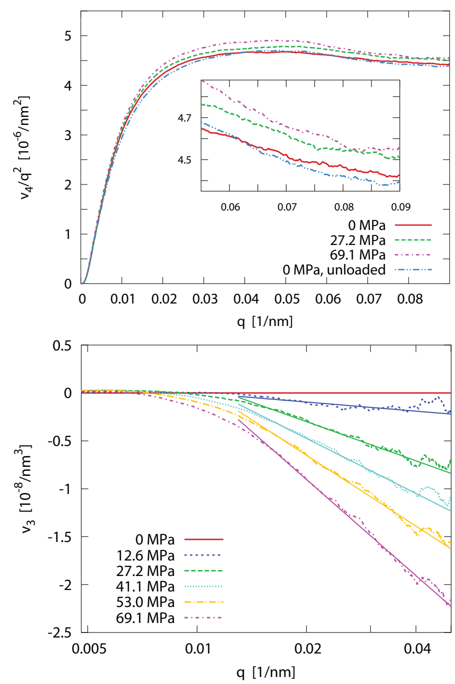

Fig. 3. The $v_4(q)/q^2$ curves measured on a sample deformed elastically up to different levels (upper box). (The pre-deformation level was 78 MPa.) The $v_3(q)$ restricted moments measured under the same conditions (lower box). For easier interpretation the $v_3(q)$ curve obtained on the unloaded sample is subtracted from the curves. Note that the horizontal axis is logarithmic.

The aim of the experimental investigations presented below was to demonstrate that the Bragg peak asymmetry varies due to external load as predicted by the theory. The measurements were performed on middle oriented Cu single crystals. The samples were plastically pre-deformed up to different compression levels. To minimize the line broadening caused by instrumental effects, the line profile measurements were performed in a double crystal diffractometer [8] using Cu K_α radiation. The intensity distributions corresponding to the (200) reflection were detected by a Bruker's position sensitive line detector. The samples were compressed in-situ by a small deformation stage, installed into the diffractometer, up to different stresses smaller than the pre-deformation level. So the samples were deformed "elastically" during the measurements.

Fig. 3. shows the restricted moments $f(q) = v_4(q)/q^2$ and $v_3(q)$ obtained on a sample deformed elastically up to different stress levels. As it is seen, the asymptotic value of $f(q)$ (according to Eq. (10) proportional to the dislocation density) increases only with a few percent with increasing load. This means, as it is expected, the dislocation density

remains nearly constant as the sample is deformed "elastically". The small increase can be attributed to the dislocation "bow out" caused by the external stress.

According to Eq. (9) the third order restricted moment of the intensity distributions become logarithmic in the asymptotic regime. Fitting a straight line to the asymptotic part the value of $\langle s\rangle$ can be determined by a quite high accuracy. (For better data analysis the $v_3(q)$ curve obtained on the unloaded sample is subtracted from the curves corresponding to deformed states.) It should be mentioned, however, that the asymmetry is rather small, so in order to get the small noise seen in Fig. 3 very precise profile measurements had to be performed. The background versus maximum intensity should be smaller than 10^{-4} [15] and the total count has to exceed 10^7.

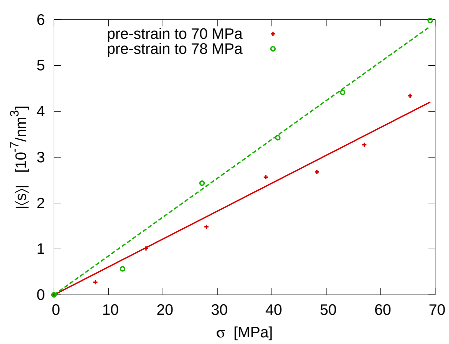

Fig. 4. The $\langle s\rangle$ versus external stress relation for two different pre-deformation levels.

The $\langle s\rangle$ versus external stress relation for two different dislocation densities are plotted in Fig. 4. The relations are linear up to the flow stress level.

The results of the line profile measurements given above are interesting to compare to the properties of the internal stress shear distribution $P_{\text{int}}(\tau)$ obtained theoretically and by discrete dislocation dynamics simulation on a system of parallel edge dislocations [16]. It should be noted that

$$\langle\tau^2\rangle = \int \tau^2 P_{\text{int}}(\tau)\mathrm{d}\tau \tag{15}$$

is proportional to the stored energy of the dislocation system, so $P_{\text{int}}(\tau)$ is a key quantity for statistical properties of dislocations. It was found by numerical calculations [16], that for large enough stress values

$$P_{\text{int}}(\tau) = C\langle\rho\rangle\frac{1}{|\tau|^3} - U\langle\rho\rangle\frac{1}{\tau|\tau|^3}, \tag{16}$$

where C is a constant, and $U = 0.85(\mu b)^2\tau_{\text{ext}}$ in which τ_{ext} is the external shear stress. So, similar to X-ray broadening the asymmetry of $P_{\text{int}}(\tau)$ increases with increasing external load.

Summing up the results obtained, it was shown that an external load applied in situ during the X-ray line profile measurement on a pre-deformed single crystal causes not only the well-known shift of the Bragg peak, but it also modifies its asymmetry. Applying the "restricted moments method" a characteristic value ($\langle s\rangle$, see Eq. (6)) determining the asymmetric asymptotic decay of the line profile can be evaluated. Due to dislocation polarization, this parameter is a measure of the variation of the dislocation-dislocation correlation function caused by an external load.

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