Mosaic Spread Analysis

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1 Mosaic Spread: Calculation

In this section, an analytical framework for a measurement of mosaic spread will be developed. Let us imagine that a sample is made up of many small domains that are tilted from the direction perpendicular to the substrate normal by some amount. A "perfect" domain is a domain that is parallel to the substrate plane. Then, we can consider a probability distribution function, $P(\alpha)$, representing a probablity of finding a domain with tilt α, which is the angle between the substrate normal and the tilted domain normal. Here, we have assumed the rotational symmetry about the substrate normal, so that the distribution does not depend on the azimuthal angle, β. The normalization condition on the probability distribution is

$$1 = \int_0^{2\pi} \mathrm{d}\beta \int_0^{\frac{\pi}{2}} \mathrm{d}\alpha \, \sin\alpha \, P(\alpha). \tag{1}$$

The object of this section is to derive the x-ray scattering structure factor including the probability distribtion. The cooridate system employed here is such that x, y, and z-axes of the zero tilt domain, that is, a domain parallel to the substrate, coincide with the lab x, y, and z-axes

First, we want to calculate the structure factor for a domain tilted by α and β, expressed in the lab coordinates. See Fig. XXX. For this, we need to express $\mathbf{q}$ in terms of . We imagine rotating the coorinates about the y-axis first, and then about the z-axis. In other words, we apply the appropriate rotation matrices to , y, and z-axes. The rotation matrix for rotaing a vector about y-axis is given by

$$\begin{pmatrix} \cos\alpha & 0 & -\sin\alpha \\ 0 & 1 & 0 \\ \sin\alpha & 0 & \cos\alpha \end{pmatrix} \tag{2}$$

and for ratating about z-axis

$$\begin{pmatrix} \cos\beta & \sin\beta & 0 \\ -\sin\beta & \cos\beta & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{3}$$

Then, what we want is

$$\hat{\mathbf{x}}' = \begin{pmatrix} \cos\beta & \sin\beta & 0 \\ -\sin\beta & \cos\beta & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \cos\alpha & 0 & -\sin\alpha \\ 0 & 1 & 0 \\ \sin\alpha & 0 & \cos\alpha \end{pmatrix} \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} = \begin{pmatrix} \cos\alpha\cos\beta \\ \cos\alpha\sin\beta \\ -\sin\alpha \end{pmatrix} \tag{4}$$

$$\hat{\mathbf{y}}' = \begin{pmatrix} \cos\beta & \sin\beta & 0 \\ -\sin\beta & \cos\beta & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \cos\alpha & 0 & -\sin\alpha \\ 0 & 1 & 0 \\ \sin\alpha & 0 & \cos\alpha \end{pmatrix} \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} = \begin{pmatrix} -\sin\beta \\ \cos\beta \\ 0 \end{pmatrix} \tag{5}$$

$$\hat{\mathbf{z}}' = \begin{pmatrix} \cos\beta & \sin\beta & 0 \\ -\sin\beta & \cos\beta & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \cos\alpha & 0 & -\sin\alpha \\ 0 & 1 & 0 \\ \sin\alpha & 0 & \cos\alpha \end{pmatrix} \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} = \begin{pmatrix} \sin\alpha\cos\beta \\ \sin\alpha\sin\beta \\ \cos\alpha \end{pmatrix} \tag{6}$$

Then, the components of $\mathbf{q}$ represented in the rotated coodinates, denoted by $\mathbf{q}'$, are the projection of $\mathbf{q}$ on x$'$, y$'$, and z$'$-axes, that is,

$$q'_x = \mathbf{q}\cdot\hat{\mathbf{x}}' = q_x\cos\alpha\cos\beta + q_y\cos\alpha\sin\beta - q_z\sin\alpha \tag{7}$$

$$q'_y = \mathbf{q}\cdot\hat{\mathbf{y}}' = -q_x\sin\beta + q_y\cos\beta \tag{8}$$

$$q'_z = \mathbf{q}\cdot\hat{\mathbf{z}}' = q_x\sin\alpha\cos\beta + q_y\sin\alpha\sin\beta + q_z\cos\alpha \tag{9}$$

The transformation rule we are looking for is

$$\cos\theta' = \frac{q'_z}{q} = \sin\theta\sin\alpha\cos(\phi-\beta) + \cos\theta\cos\alpha \tag{10}$$

and

$$\tan\phi' = \frac{q'_y}{q'_x} = \frac{\sin\theta\sin(\phi-\beta)}{\sin\theta\cos\alpha\cos(\phi-\beta) - \cos\theta\sin\alpha} \tag{11}$$

The structure factor of the tilted domain in the lab coordinates is simply given by $S(\mathbf{q}') = S(q, \theta', \phi')$. Summing over all the domains, we get for the total structure factor

$$S_M(q, \theta, \phi) = \int_0^{2\pi} \mathrm{d}\beta \int_0^{\frac{\pi}{2}} \mathrm{d}\alpha\, \sin\alpha\, S(q, \theta', \phi')\, P(\alpha) \tag{12}$$

with Eq. (10) and Eq. (11).

For $\theta = 0$, $\theta' = \alpha$ and $\phi' = 0$ or π, so we have

$$\begin{aligned} S_M(q, 0) &\sim \int_0^{\frac{\pi}{2}} \mathrm{d}\alpha\, \sin\alpha\, S(q, \alpha, 0)\, P(\alpha) + \int_0^{\frac{\pi}{2}} \mathrm{d}\alpha\, \sin\alpha\, S(q, \alpha, \pi)\, P(\alpha) \quad (13)\\ &= \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \mathrm{d}\alpha\, \sin\alpha\, S(q, \alpha)\, P(\alpha) \quad (14) \end{aligned}$$

if we understand $S(q, \alpha)$ to be the structure factor on the (q_r,q_z) plane. This shows that $S_M(q, 0)$ is equal to the convolution of the distribution function and the original structure factor. In general, however, $S_M(q, \theta)$ is not a convolution of the distribution function and the structure factor.

Given Eqs. (10), (11), and (12), we want to show that mosaic spread acts as one dimensional convolution in the x-ray structure factor:

$$S_M(q, \theta) = \int_{-\pi}^{\pi} \mathrm{d}\alpha\, S(q, \theta - \alpha)\, P(\alpha) \tag{15}$$

The structure factor representing Bragg peaks in the spherical coordinates are written as

$$S(q, \theta, \phi) \sim \frac{\delta(q - \frac{2\pi h}{D})}{q^2} \delta(\cos\theta - 1)\delta(\phi) \tag{16}$$

where $\delta(x)$ is the Dirac delta function. Plugging Eq. (16) in Eq. (12), we obtain $\phi - \beta = 0$. Using this condition, we get

$$S_M(q, \theta) \sim \frac{\delta(q - \frac{2\pi h}{D})}{q^2} P(\theta) \sin\theta, \tag{17}$$

which shows that we can directly measure the probability distribution experimentally by looking at the intensity along $q = 2\pi h/D$. In the next section, we will discuss the relevant experimental techniques.

2 Mosaic Spread: Experiment

In this section, we discuss experimental procedures to probe appropriate q-space to measure the mosaic distribution, $P(\alpha)$. In our setup, the angle of incidence between the beam and substrate, denoted by ω, can be varied. A conventional method to measure mosaicity distribution is a rocking scan, where one measures the integrated intensity of a given Bragg peak as a function of ω with a fixed detector position. In a non-conventional method called ring analysis, one measures the intensity as a function of η on a two dimensional detector. First, we want to show that the two methods mentioned above in fact measure the mosaicity disitribution and therefore are equivalent to each other.

Let ω be the angle of incidence, 2θ be the total scattering angle, p_x be the pixel number in the horizontal direction, p_z be the pixel number in the vertical direction, *eta* be the angle measured from the p_z-axis on the detector. Δp is 0.07113 mm/pixel. $q_x = \mathbf{q}\cdot\hat{\mathbf{x}}$, $q_y = \mathbf{q}\cdot\hat{\mathbf{y}}$, and $q_z = \mathbf{q}\cdot\hat{\mathbf{z}}$, where $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$ are all defined on the sample space. This means that $\hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$ both rotate as ω is varied while $\hat{\mathbf{x}}$ is always perpendicular to the beam. What we need is a set of transformation rules for going from the detector space (pixels) to the sample q-space. With them, we would know how to trace out a line on the detector in order to measure the mosaicity distribution.

The incoming and outgoing wavevectors of the x-ray beam in Fig. XXX are given by

$$\mathbf{k}_{\text{in}} = \frac{2\pi}{\lambda}\hat{\mathbf{y}}, \quad \mathbf{k}_{\text{out}} = \frac{2\pi}{\lambda}\left(\sin 2\theta\cos\phi\,\hat{\mathbf{x}} + \cos 2\theta\,\hat{\mathbf{y}} + \sin 2\theta\sin\phi\,\hat{\mathbf{z}}\right), \tag{18}$$

where λ is the wavelength of x-ray. The scattering vector is the difference between $\mathbf{k}_{\text{in}}$ and $\mathbf{k}_{\text{out}}$,

$$\begin{aligned}\mathbf{q} &= \mathbf{k}_{\text{out}} - \mathbf{k}_{\text{in}} \\ &= q\left(\cos\theta\cos\phi\,\hat{\mathbf{x}} - \sin\theta\,\hat{\mathbf{y}} + \cos\theta\sin\phi\,\hat{\mathbf{z}}\right),\end{aligned} \tag{19}$$

where $q = 4\pi\sin\theta/\lambda$ is the magnitude of the scattering vector. When the sample is rotated by ω about the x-axis in the clockwise direction as shown in Fig. XXX, the sample coodinates written in terms of the lab coordinates are

$$\hat{\mathbf{e}}_{\mathbf{x}} = \hat{\mathbf{x}}, \quad \hat{\mathbf{e}}_{\mathbf{y}} = \cos\omega\,\hat{\mathbf{y}} + \sin\omega\,\hat{\mathbf{z}}, \quad \hat{\mathbf{e}}_{\mathbf{z}} = -\sin\omega\,\hat{\mathbf{y}} + \cos\omega\,\hat{\mathbf{z}}. \tag{20}$$

From Eq. (19) and Eq. (20), we find the projection of $\mathbf{q}$ on the sample coordinates to be

$$q_x = \mathbf{q} \cdot \hat{\mathbf{e}}_{\mathbf{x}} = q \cos\theta \cos\phi \tag{21}$$

$$q_y = \mathbf{q} \cdot \hat{\mathbf{e}}_{\mathbf{y}} = q \left(-\sin\theta \cos\omega + \cos\theta \sin\phi \sin\omega\right) \tag{22}$$

$$q_z = \mathbf{q} \cdot \hat{\mathbf{e}}_{\mathbf{z}} = q \left(\sin\theta \sin\omega + \cos\theta \sin\phi \cos\omega\right). \tag{23}$$

With respect to the beam, the position on the detector is given by

$$X = S \tan 2\theta \cos\phi, \quad Z = S \tan 2\theta \sin\phi. \tag{24}$$

The pixels on the detector are directly proportional to X and Z. Thus, these equations define the transformation rules from the detector space to the sample q-space and vice versa.

In terms of these coordinates, in the rocking scan, $\phi = \pi/2$ and $q = 2\pi h/D$ while ω is varied about θ_B, where θ_B is the Bragg angle for a Bragg peak that is focused on. Using $q = 4\pi \sin\theta/\lambda$, we recover the Bragg condition, $2D \sin\theta = h\lambda$. Plugging $\phi = \pi/2$ in Eq. (21), (22), and (23),