Dynamical-Theory computation of Multiple-Beam Diffraction

Coordinate System (CS):

Fig. 1(a) shows the Cartesian CS for computing **Multiple-Beam Diffraction** (MBD), where the **Z**-axis is parallel to the diffraction vector **G** of the **Primary Reflection**, **X**-axis is the **Horizontal Reference Direction** the user must choose, and $\mathbf{Y} = \mathbf{Z} \times \mathbf{X}$. The direction of the incident beam is described by the **diffraction angle** θ (relative to the Bragg plane) and the **azimuthal angle** Φ [relative to **X**-axis, $\Phi > 0$ along the arrow direction of Φ (toward $+\mathbf{Y}$) in Fig. 1(a), and $\Phi < 0$ along the opposite direction]. So the incident wavevector is $\mathbf{K}_0 = K(\cos\Phi\cos\theta\,\hat{\mathbf{x}} + \sin\Phi\cos\theta\,\hat{\mathbf{y}} - \sin\theta\,\hat{\mathbf{z}})$, and the diffracted wavevector of the primary reflection is $\mathbf{K}_G \approx K(\cos\Phi\cos\theta\,\hat{\mathbf{x}} + \sin\Phi\cos\theta\,\hat{\mathbf{y}} + \sin\theta\,\hat{\mathbf{z}})$, where $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, $\hat{\mathbf{z}}$ are unit vectors along X, Y, Z, respectively, and $K = 1/\lambda$ (λ the incident X-ray wavelength).

The **diffracting lattice planes**, also called the Bragg planes, are perpendicular to the **diffraction vector**. If the **crystal surface** is parallel to the Bragg plane, the configuration is called the **symmetric Bragg case** (of two-beam diffraction). In general, the crystal surface has an **offcut angle** with respect to the Bragg planes (of the primary reflection), denoted by Ω_{off} in Fig. 1(b). Meanwhile, the **offcut direction** is described by the azimuthal angle Φ_{off}. Note that the offcut can dramatically change the diffraction properties. For example, Fig. 1(c) shows the diffraction geometry for $\Phi_{\text{off}} = 180°$ and $\Omega_{\text{off}} = 90°$, which becomes a **symmetric Laue case** (for a finite crystal thickness).

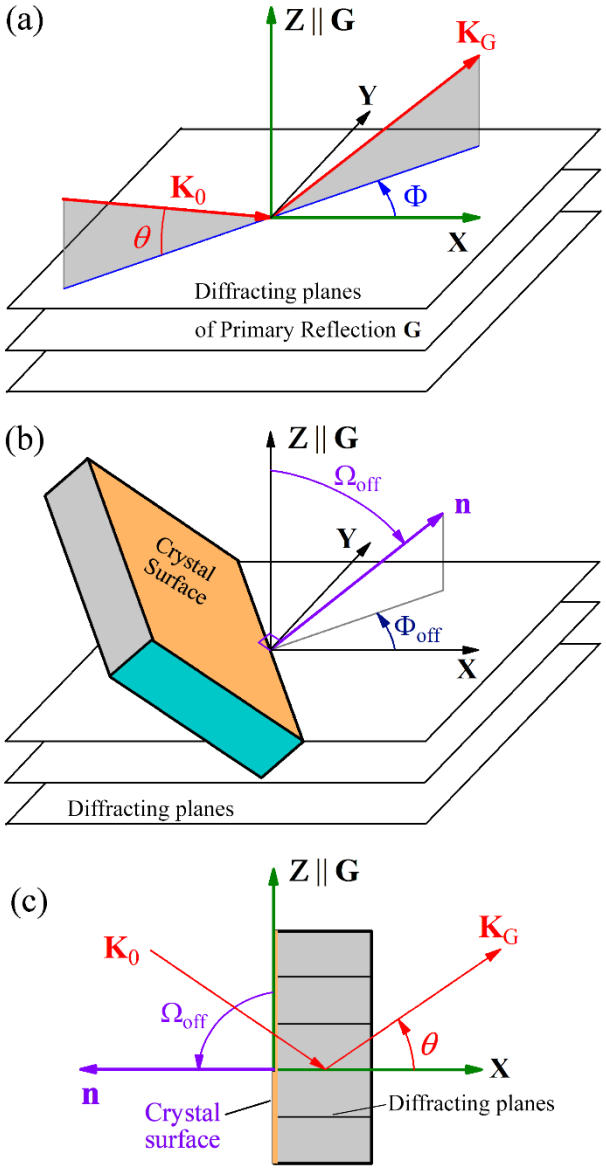

Fig. 1. (a) The CS XYZ for calculating MBD. (b) Description of the Crystal Surface in the CS by the Ω_{offcut} and Φ_{off} angles. **n** is the crystal surface normal. (c) The symmetric Laue case with $\Phi_{\text{off}} = 180°$ and $\Omega_{\text{off}} = 90°$.

Steps to Set Parameters:

To run the program, you first set the **Photon Energy**. Then import a **crystal structure** and select the **primary reflection** (so **Z**-axis is determined), followed by choosing the **Horizontal Reference Direction** (hkl) to determine the **X**-axis, *i.e.* **X** is parallel to the *reciprocal lattice vector* $h\mathbf{a}^* + k\mathbf{b}^* + h\mathbf{c}^*$ in *reciprocal space*, NOT in real space.[1]

Fig. 1(a) is based on the **two-beam diffraction** geometry (for 000 and **G**). When θ is fixed at the Bragg angle θ_B while Φ is varied, reflection **G** is always activated. However, at discrete angles Φ, the incident beam may also satisfy the Bragg conditions of other reflections in 3D space, giving rise to MBD. In the program, in addition to the first reflection $\mathbf{G}_0 = 000$ and the second reflection $\mathbf{G}_1 = \mathbf{G}$, the button **Add Reflection** allows you to add a third reflection $\mathbf{G}_2$ (here we use **0-based indices**). Then the program calculates the Φ angles (usually two, denoted by Φ_{3BD} here) that fulfill the **three-beam**

[1] For cubic structures, $h\mathbf{a}^* + k\mathbf{b}^* + h\mathbf{c}^*$ in reciprocal space is parallel to vector $h\mathbf{a} + k\mathbf{b} + h\mathbf{c}$ in *real space*, but note that for non-cubic structures, these two vectors (directions) may NOT be parallel.

diffraction condition. You can select either angle (by clicking the **Set Azimuth Angle of 3-Beam Diffraction** or **2nd** buttons) as the **Principal Azimuth angle of Incident Beam**. Around (Φ_{3BD}, θ_B) you can then calculate the 3-beam **diffraction intensity contour** in terms of (Φ, θ) of the Bragg planes --- NOT the crystal surface! In fact, you can add more reflections and you may choose any Principal Azimuth angle of Incident Beam to compute MBD. If the Bragg condition of a reflection you included is not satisfied, it would not affect the computing results! If none of the additional reflections is effective, the program gives you the conventional **two-beam diffraction** (000 and **G**) results! Therefore, you can add as many reflections as you like, even irrelevant reflections!---but more reflections could slow down the computing.

You can click **Calculate or Import Chi Components** to obtain the complex **Chi (χ) components** (proportional to the **structure factors**). For N-beam diffraction, the Chi components form a $N \times N$ matrix

$$\begin{pmatrix} \chi_{0-0} & \chi_{0-1} & \cdots & \chi_{0-(N-1)} \\ \chi_{1-0} & \chi_{1-1} & \cdots & \chi_{1-(N-1)} \\ & & \vdots & \\ \chi_{(N-1)-0} & \chi_{(N-1)-1} & \cdots & \chi_{(N-1)-(N-1)} \end{pmatrix}$$

where the 0-based indices $(m-n)$ of each component indicate its corresponding diffraction vector $\mathbf{G}_m - \mathbf{G}_n$ (with $\mathbf{G}_0$ = 000, $\mathbf{G}_1 = \mathbf{G}$, $\mathbf{G}_3$, $\mathbf{G}_4$, … $\mathbf{G}_{N-1}$ being the involved reflections). The program can automatically calculate the χ components, or you can import your own χ data from a text file.

The **polarization state** is always defined with respect to the two-beam diffraction configuration of Fig. 1(a). For Sigma (σ) polarization, the magnetic field **H** of the incident beam lies in the plane containing both $\mathbf{K}_0$ and **G** and meanwhile is perpendicular to $\mathbf{K}_0$, i.e. $\mathbf{H}^\sigma = H^\sigma(\cos\Phi\sin\theta\,\hat{\mathbf{x}} + \sin\Phi\sin\theta\,\hat{\mathbf{y}} + \cos\theta\hat{\mathbf{z}})$. For Pi ($\pi$) polarization, **H** is perpendicular to both $\mathbf{K}_0$ and **G**, i.e., $\mathbf{H}^\pi = H^\pi(-\sin\Phi\,\hat{\mathbf{x}} + \cos\Phi\,\hat{\mathbf{y}})$. Note that it is the same as $\mathbf{E}^\sigma = E^\sigma(-\sin\Phi\,\hat{\mathbf{x}} + \cos\Phi\,\hat{\mathbf{y}})$ and $\mathbf{E}^\pi = E^\pi(\cos\Phi\sin\theta\,\hat{\mathbf{x}} + \sin\Phi\sin\theta\,\hat{\mathbf{y}} + \cos\theta\hat{\mathbf{z}})$ as described in the conventional dynamical theory. In the MBD program, the **Polarization angle** allows you to define a mixed polarization state. When the polarization angle is 0, it corresponds to pure Sigma polarization. When the polarization angle is 90°, it corresponds to pure Pi polarization. Between 0 and 90°, it is mixed polarization, and the Polarization angle is the angle between the wave amplitude and the "Sigma Axis". Currently only **linear polarization** of the incident beam is implemented. **Elliptical polarization** of incidence can be easily implemented in the future if users request. (Or you can do it yourself. First calculate the Sigma polarization results, and then separately calculate another set of data for Pi polarization. Then during your own data processing, you may add a phase factor to the output of the Pi wave amplitudes. You may also need to add a magnitude factor to the Pi results based on the strength ratio of your incident Sigma and Pi components of your elliptical incident wave. Then add the two results together.)

The parameter ΔE allows you to compute MBD by slightly deviating the **X-ray energy** from the Bragg **Photon Energy** you specified at the beginning. The $\Delta\theta$ and $\Delta\Phi$ scanning ranges allow you to calculate the 2D MBD intensity contour as a function of (Φ, θ). If you want a 1D Darwin curve in terms of only θ (or Φ), you can simply ***set the points of the other angular parameter,*** Φ (or θ), ***to be ONE*** **(1)**. Then the other parameter is the starting value of the range and will be contestant. The "Si/Ge/C structure" choice is for the diamond structure (Si, Ge, diamond). This choice is automatically checked if the crystal name is one of "Si", "Ge", "C", "Silicon", "Germanium", "Diamond". You can manually check/de-check it. Checking this choice can significantly speed up the computing for Si/Ge/C crystals.

The 3D vectorial dynamical theory of MBD used in the program is based on the **Fourier coupled-wave diffraction theory (FCWDT)** by X.-R. Huang, et al., *Phys. Rev. A* **87**, 063828 (2013) (Section IV). In this theory, each involved reflection $\mathbf{G}_m$ activates a diffracted wave $\mathbf{E}_m^R \exp(-i\mathbf{K}_m^R \cdot \mathbf{r})$ above the crystal surface [with $\mathbf{E}_0^R \exp(-i\mathbf{K}_0^R \cdot \mathbf{r})$ being the specular reflection of the surface] $(m = 0, 1, 2 \cdots, N-1)$. If the crystal is a parallel plate with a finite thickness, there are another N waves $\mathbf{E}_m^T \exp(-i\mathbf{K}_m^T \cdot \mathbf{r})$ below the exit surface [with $\mathbf{E}_0^T \exp(-i\mathbf{K}_0^T \cdot \mathbf{r})$ being the forward transmitted wave], which is true for both the Bragg and Laue cases.

For simplicity, this program does NOT provide a graphic interface for visualizing the MBD results. Instead, it always opens a file for you to export the computed results. Afterwards, you can use third-party software (e.g. ORIGIN) to plot the contour or curves using the file. Usually, you are interested in the reflectivity of the primary reflection ($\propto \left|\mathbf{E}_1^R\right|^2$ for the Bragg case or $\propto \left|\mathbf{E}_1^T\right|^2$ for the Laue case). But the program allows you to export the reflectivity of any reflection $m = 0, 1, 2, \cdots, N-1$. The default format of the output file has **three columns**:

$$\Delta\Phi,\ \Delta\theta,\ R_m$$

where the unit of $\Delta\Phi$ and $\Delta\theta$ can be either **arcsec** or **μrad**, and R_m is the reflectivity of the mth reflection (for $m = 0, 1, 2, \cdots, N-1$). If you use **Origin**, you can import the three-column data. Then select the third column, C(Y), and use Menu "Column → Set As" to set this column as Z. Then use Menu "Plot → Contour/Heat Map → Color Fill" to plot the contour. By default, Origin may plot the contour with "Speed Mode is On". You can use Menu "Graph → Speed Mode → Open Dialog" to turn off the Speed Mode.

For semi-infinite crystals, you can only calculate the reflectivity on the Entrance Crystal Surface. For finite-thickness crystals, you may calculate the **transmission reflectivity** on the **Exit Crystal Surface Side**. For the **Laue transmission case** of a beam [such as Fig. 1(c)], only the transmission reflectivity has noticeable values in general.

The program also allows you to export the amplitudes and wavevectors of the mth reflection. For this option, you can choose the complete format of the output file with the following **columns**:

$\Delta\Phi,\ \Delta\theta,\ R_m,\ H_m^\sigma$ real part, H_m^σ imaginary part, H_m^π real, H_m^π imag, K_{mx}, K_{my}, K_{mz}

Here K_{mx}, K_{my}, K_{mz} are the components of the unit vector $\widehat{\mathbf{K}}_m$ along the mth diffracted wavevector $\mathbf{K}_m$ in the XYZ CS of Fig. 1(a). The wave amplitude (for unit incidence) is expressed by its two components H_m^σ and H_m^π along the two orthogonal directions $\mathbf{n}^\sigma = \widehat{\mathbf{K}}_0 \times \widehat{\mathbf{K}}_m$ and $\mathbf{n}^\pi = \widehat{\mathbf{K}}_m \times \mathbf{n}^\sigma$ that are perpendicular to $\mathbf{K}_m$. H_m^σ and H_m^π are complex numbers, and you can also export H_m^σ and H_m^π in the phase form (the default setting) consisting of the **module** and **phase** (in degree). From the wave amplitudes (including the forward transmitted wave with $m = 0$ in the transmission geometry) you may analyze the **polarization change**, the formation of **elliptical/circular polarization**, and the **phase change** in MBD. Note again the output polarization is for the magnetic field **H**, not for the electric field **E**.

Divergent/polychromatic Incident Beam

The above computation is based on a plane-wave incident beam, where you calculated the reflectivity as a function of $(\Delta\Phi, \Delta\theta)$. At the bottom, by clicking "**Activate Convolution Panel**", the program also allows you to use an incident beam with divergence along both the Φ and θ directions and a finite (FWHM) spectral bandwidth (Gaussian shape). Then for each point $(\Delta\Phi, \Delta\theta)$ --- the same parameters in the above Plane-Wave panel --- the program makes multiple integration over the two **Beam Divergence Ranges** and the **energy Integration Range**. Therefore, the multiple integration could be extremely slow, and may take hours to days. For testing the convergence, you may limit the integral over small ranges and make a 1D scan of $\Delta\Phi$ or $\Delta\theta$ --- don't do the 2D $(\Delta\Phi, \Delta\theta)$ mapping during testing. Then you can test each parameter of the Range or Points in the Convolution Panel. When the results show no noticeable difference, you reach the convergence for this parameter. After you think all the parameters reach the convergence requirements, you may make full scan of $\Delta\Phi$ and/or $\Delta\theta$.

Reflections with Azimuth Angles (Φ) close to the Principal Azimuth Angle of Incidence

The right panel of the MBD program is "**Check Reflections with Azimuth Angles close to current Principal Azimuth Angle of Incidence**". It searches, in the vicinity of the current **Principal Azimuth Angle** (you must set), if there

are other reflections with their Bragg conditions nearly satisfied. For example, at E = 8 keV, Primary Reflection Si 004, Horizontal Reflection Direction 100. The Φ angle of Reflection 111 is $-38°$. If you set the Principal Azimuth Angle to be $-38°$, you would think that now it is a 000-004-111 three-beam diffraction condition. However, if you check other reflections in the right panel, you can find that reflection 113 also has the same Φ angle ($\Delta\Phi = 0$). Therefore, this is, in fact, a **000-004-111-113 four-beam diffraction** configuration.

You may include the searched reflections in the MBD computing in the list of the left panel if there ΔΦ values are very close to zero. Note that some of the searched reflections may be **forbidden reflections**. However, a three-beam diffraction (000-$\mathbf{G}_1$-$\mathbf{G}_2$) process actually involves a hidden diffraction channel $\mathbf{G}_{\text{hidden}} = \mathbf{G}_1 - \mathbf{G}_2$. The three-beam diffraction process still occurs if only two of the three reflections $\mathbf{G}_{\text{hidden}}$, $\mathbf{G}_1$, $\mathbf{G}_2$ are allowed reflections. For example, Si 000-111-002 three-beam diffraction exists because it involves reflections 111, 002, and -1-11. Reflections 111 and -1-11 are allowed reflections although 002 direct reflection is forbidden, i.e., the diffraction process is a detour reaching 002 through 111 and -1-11.

The button "**Multiple-Beam Diffraction Lines (Monochromator Glitches) in Azimuth-Energy Map...**" allows you to activate the "**Multiple-Beam Diffraction Lines (Monochromator Glitches) in Azimuth-Energy Map**" that can provide a full Φ-*E* map of the MBD lines *based on the current crystal and configuration*, see the following section.

MBD lines in Azimuth-Energy ($\Phi - E$) Coordinate System

This program provides fast mapping of **multiple-beam diffraction lines**, also called **Monochromator Glitches** in the EXAFS community, in the Azimuth-Energy space. It particularly includes those **forbidden reflections with allowed detour reflections**, such as the above Si 000-111-002 three-beam diffraction case, i.e., for a three-beam diffraction case 000-$\mathbf{G}_1$-$\mathbf{G}_2$, if two of the three involved reflections $\mathbf{G}_1$, $\mathbf{G}_2$, $\mathbf{G}_{\text{hidden}} = \mathbf{G}_1 - \mathbf{G}_2$ are allowed reflections, the three-beam diffraction conditions are fulfilled [see X.-R. Huang *et al*., **Appl. Phys. Lett. 105**, 181903 (2014)**; J. Appl. Cryst. 47**, 1716-1721 (2014)]. For Si/Ge/C structures, the program uses the diamond structure rule to judge if a reflection hkl is allowed (h, k, l are all even numbers with $h + k + l = 4n$, or h, k, l are all odd numbers) or forbidden. For general crystals, the program calculates the χ **factor** of each reflection, and if its absolute value is less than a **threshold value** (default is 10^{-8}), the reflection is treated as a forbidden reflection. Note that in general, $\chi \sim 10^{-6} - 10^{-5}$ for allowed reflections. The computing algorithm has been highly optimized so that the computing is very fast for any crystals. You can change the **threshold value**, or you can set it to zero if you want to ignore this restriction.

The "**Save Line…**" function allows you to save to a file the **Glitch points** on any vertical line Φ = constant. This is useful for selecting lateral directions to avoid or optimize glitches during X-ray crystal monochromator designing.

You can click an MBD line to see its reflection index. For Si/Ge/C structure, different reflections may have identical lines overlapped together. So, if you click a line more than once, you may get different reflection indices. You can also allow the program to print the reflection index of each line. The index will be plotted at random positions on the line. So you can let the program print/hide the indices for more times to randomly change the label positions so that you can clearly identify each line when the lines or labels are overlapped.