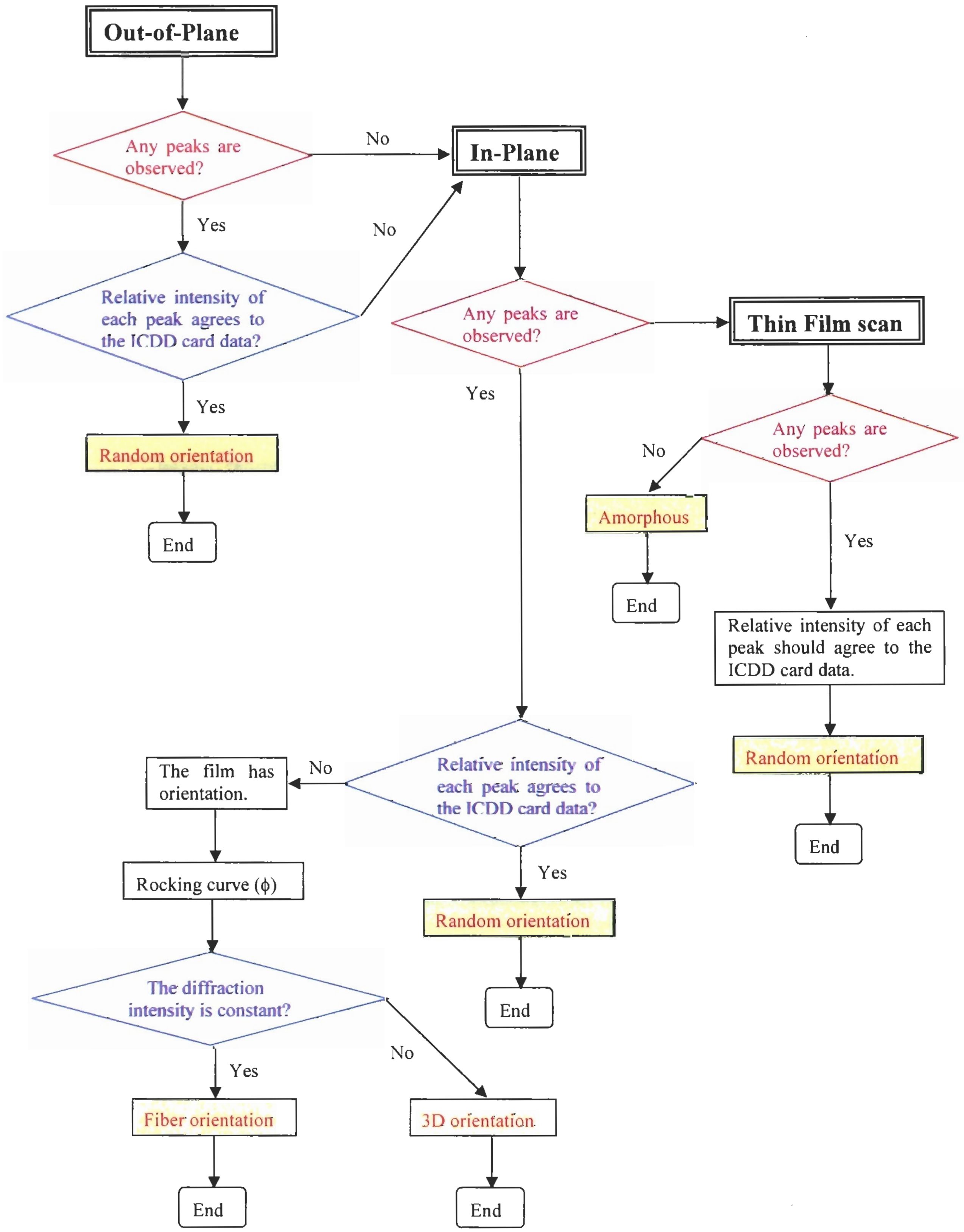
1. X-ray diffraction

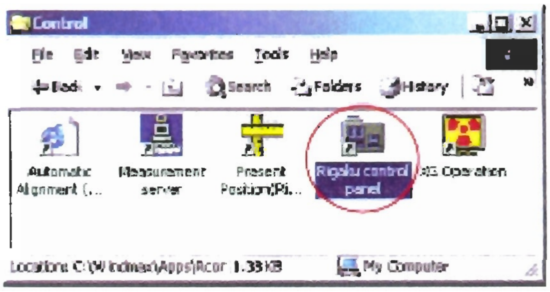
1-1 Flow chart

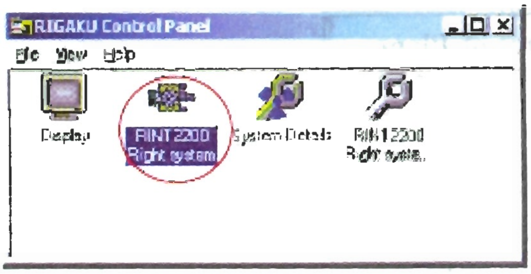


1-2 X-ray diffraction Measurement

1-2-1 Setting up the geometry system

Open “Rigaku control panel” from the Rigaku folder. Double click “RINT 2200 Rigaku system. ”





Select following items;

Attachment :

Attachment for thin film/ UltimaIII

Incident monochro:

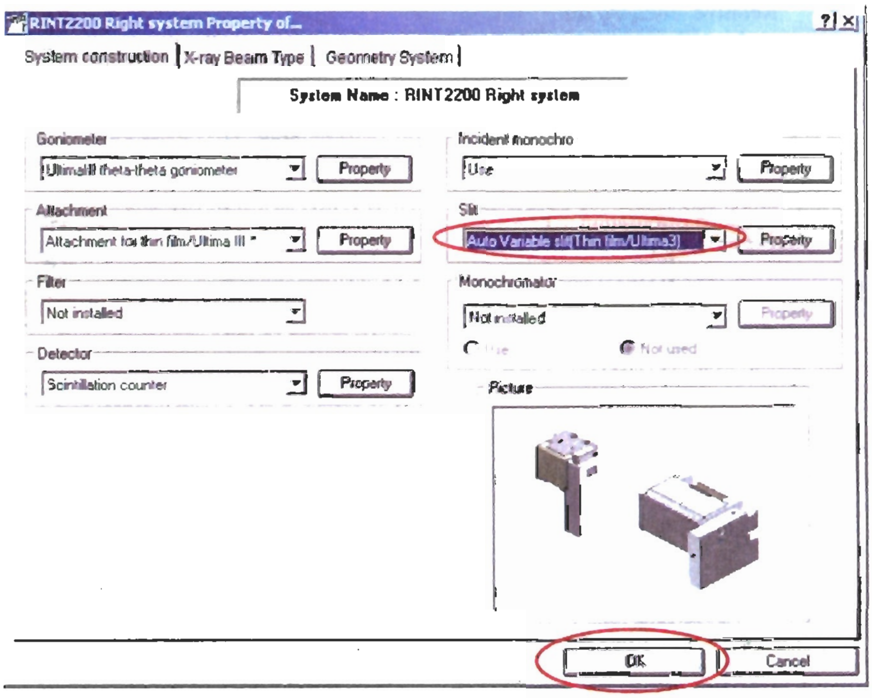
Use

Slit:

Auto Valiable slit(Thin film/ UltimaIII)

Monochrometer:

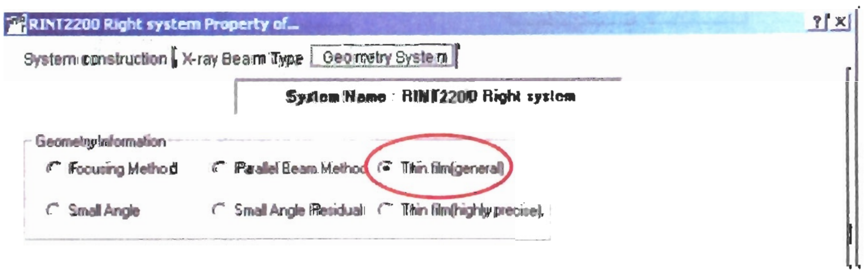
Not installed



Geometry Information:

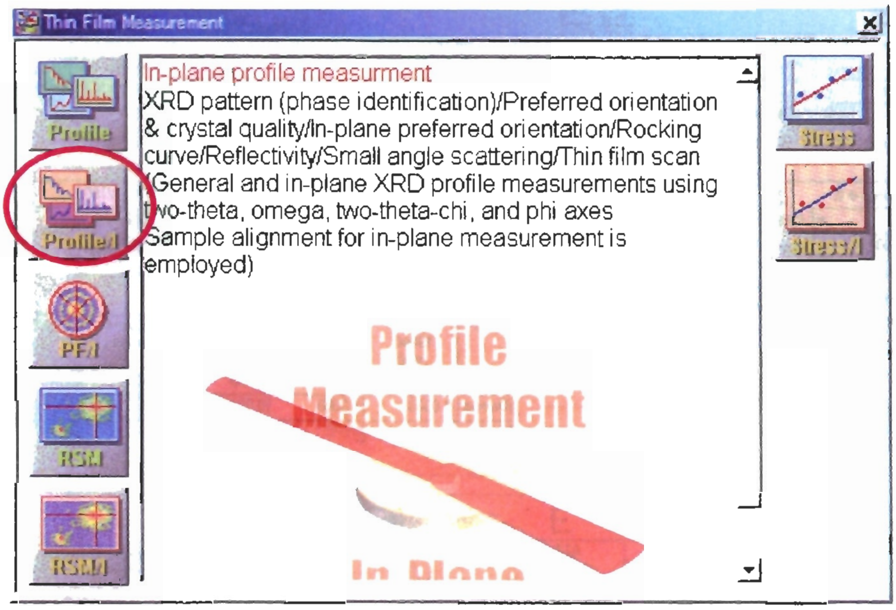
Thin film (general) or

Thin film (highly precise)

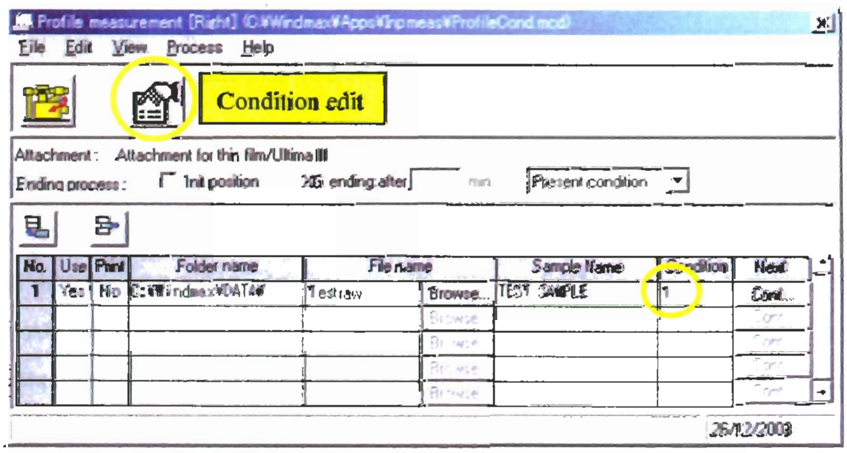


1-2-2 Out-of-Plane Measurement

Open [Start] – [Programs] - [Rigaku] – [Rigaku Measurement] – [Thin Film Meas. Menu.]



Select [Profile/In-Plane] measurement.

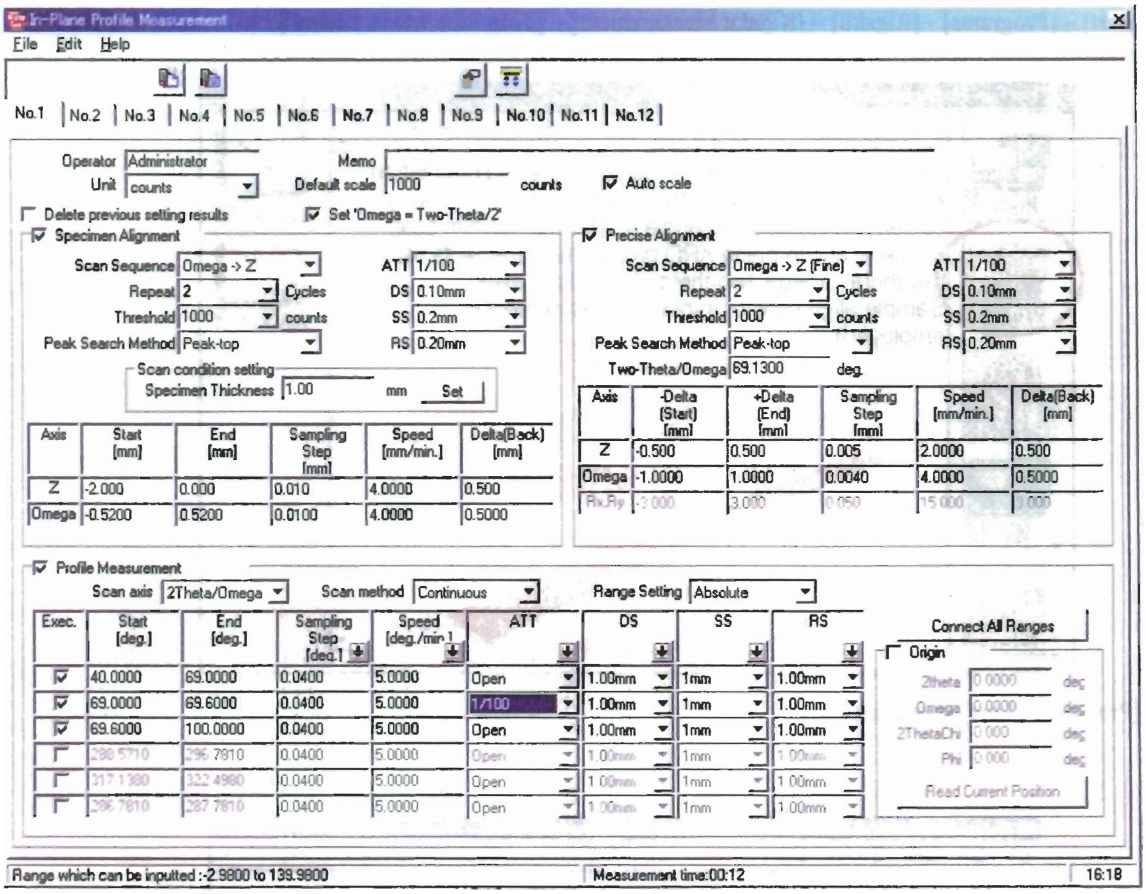


Input the folder name, the file name and the sample name. Switch "Use" to "Yes" (otherwise the measurement will not be performed!) To print the raw data (measured profile and the scanning condition) after the measurement, switch "Print" to "Yes."

If you want to perform a series of scans, click "No. " and "copy" button; the measurement condition will be copied. To remove conditions, use "Remove" button.

To edit measurement conditions, click on the "condition edit" button or double-click on the number input in [Condition.]

An example of scanning condition for Out-of-Plane measurements in as follows;

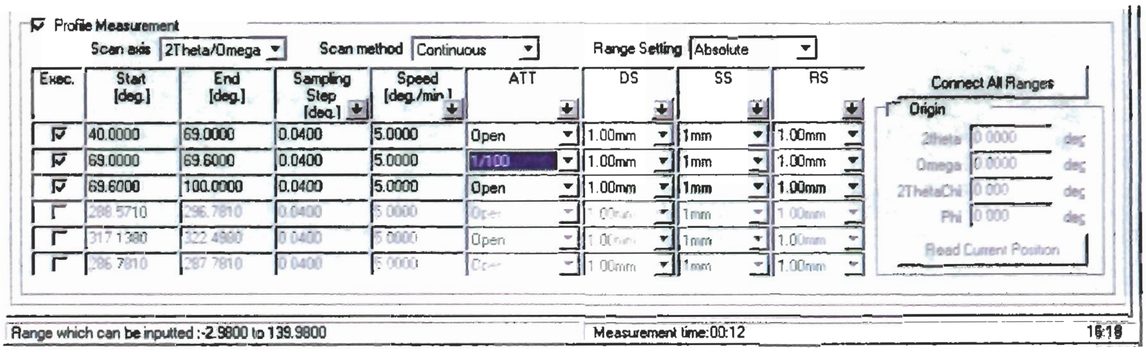


☆Specimen alignment

"Specimen alignment" has to be performed before Out-of-Plane measurement. Input "Specimen Thickness" and click "Set." The recommended scanning conditions of Z and Omega axes are input automatically. "Precise Alignment" is used when users need to be sure the direction of lattice plane observed with 2θ/ω scanning. When the critical angle for total reflection is input to "Two Theta/Omega," the lattice planes parallel to the sample surface are observed; when the diffraction angle of the substrate is input, the lattice planes parallel to the substrate's lattice planes are observed.

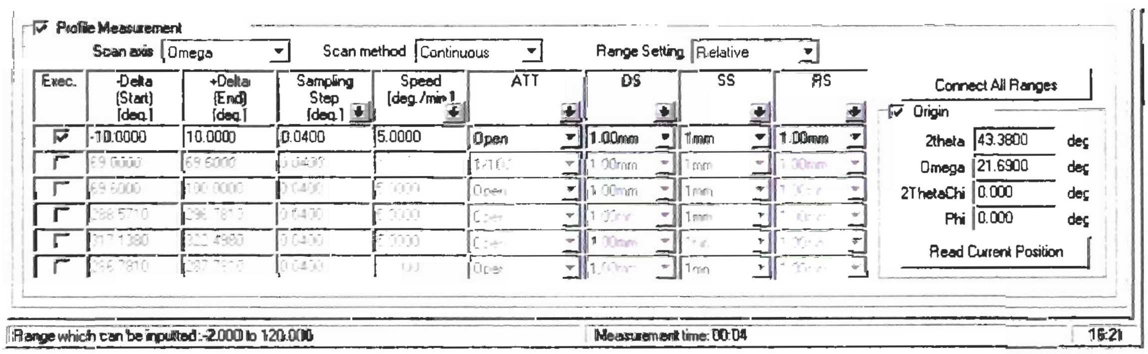
☆Out-of-Plane measurement (2Theta/Omega scan)

"2theta/omega" is selected as "Scan axis." The scanning range is determined by referring the ICDD card data. The sampling step should be 1/5 ~ 1/10 of a diffraction peak. Switch the attenuator (ATT) at the middle of the scanning range according to the peak intensity, if necessary.



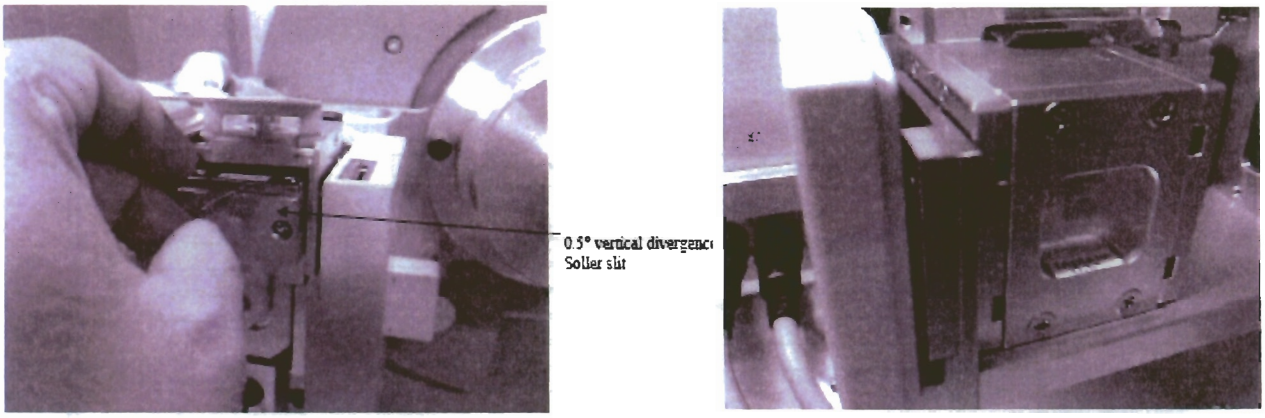
☆Rocking curve (Omega)

"Omega" is selected as "Scan axis." Input the 2theta angle of the diffraction peak and the half value into "2Theta" and "Omega" in "Origin", respectively.

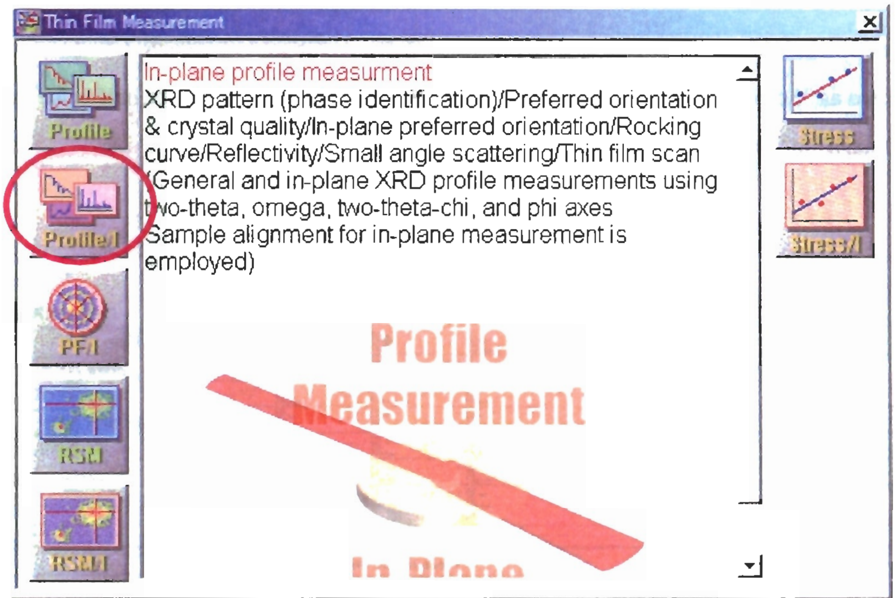


1-2-3 In-Plane Measurement

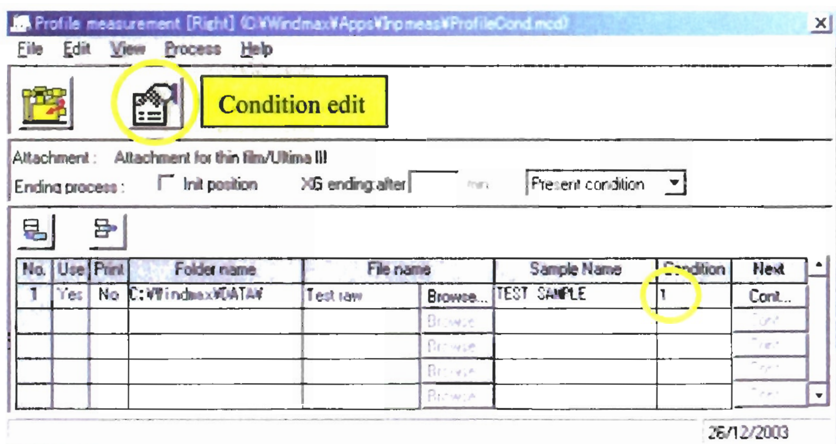
Before starting the measurements, vertical divergent Soller slits are installed.



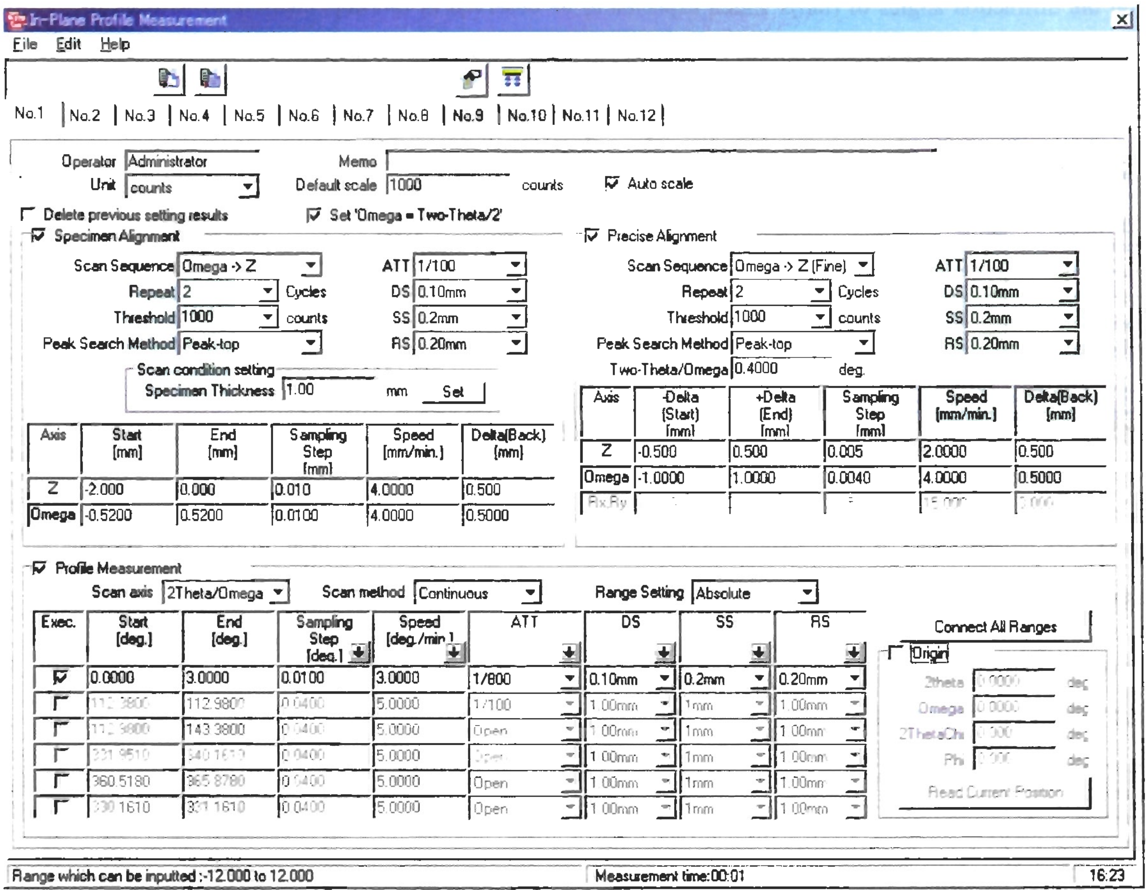
Open [Start] – [Programs] - [Rigaku] – [Rigaku Measurement] – [Thin Film Meas. Menu.]



Select [Profile/In-Plane] measurement.



This is an example of scanning condition for preliminary scan of In-Plane measurement.



☆Specimen alignment

For In-Plane measurement, both “Specimen alignment” and “Precise Alignment” has to be performed before measurement. The critical angle for total reflection is input to “Two-theta/Omega”; this procedure makes the incident angle (Omega) constant for any phi angle.

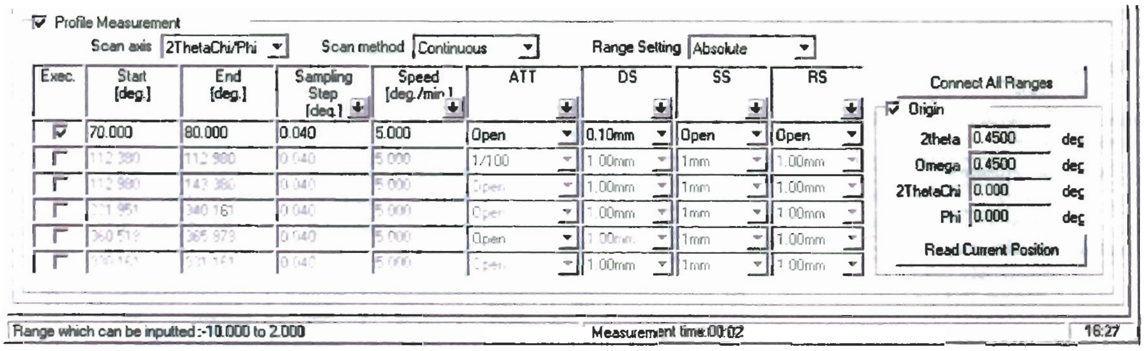
☆Preliminary measurement

①Examination of critical angle

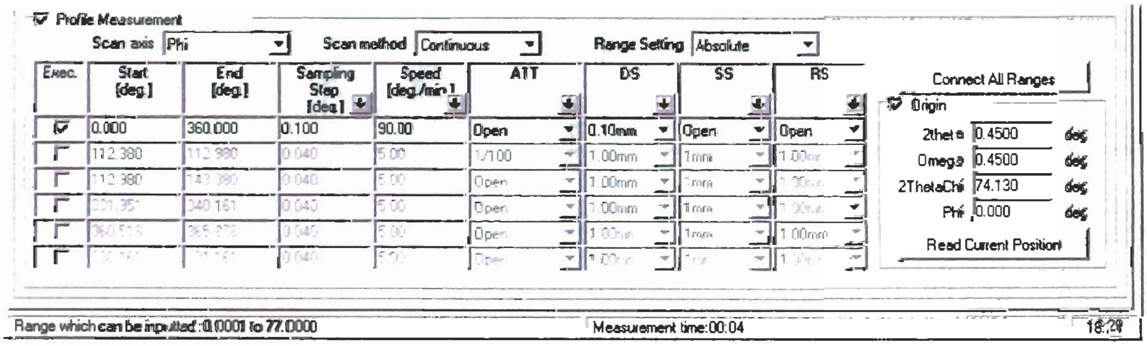
To observe the diffraction from the samples, incident x-rays have to penetrate to the sample. When the incident angle is smaller than the critical angle for total reflection, the penetration depth is just few nanometers and sometimes diffracted intensity becomes extremely low. To estimate the critical angle, perform 2Theta/Omega scan around critical angle (0~3 deg.) Read critical angle θ_c and input the value into “2Theta” and “Omega” in “Origin”.

②Finding peaks

Check on the diffraction angles of (hkl)s which expected to be observed in In-Plane measurements from the results of Out-of-Plane measurement. Perform 2ThetaChi/Phi scanning around the angles.

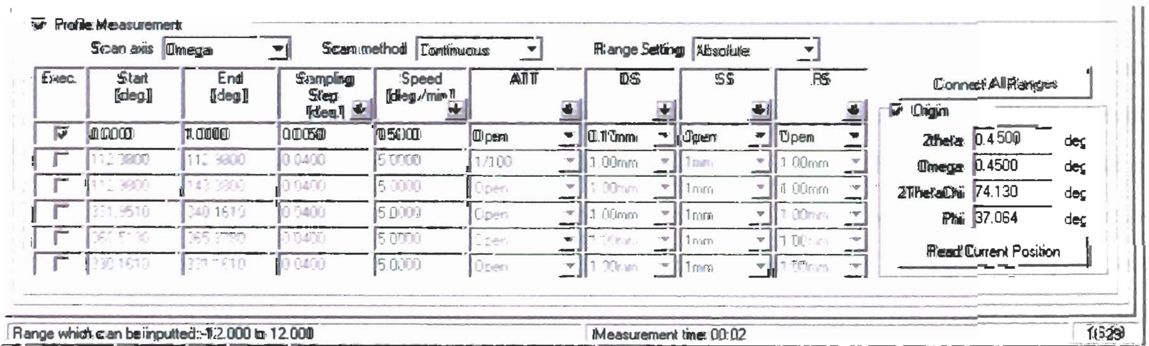


If any peaks are not observed, it is supposed that the film is three-dimensionally oriented. In this case, try Phi scan with fixing 2ThetaChi angle at a diffraction angle estimated above.



③Estimation of incident angle

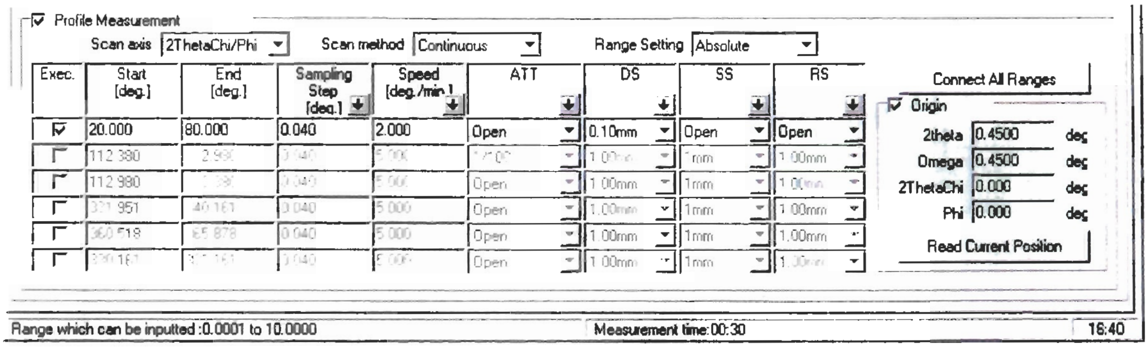
Input the peak position into “2ThetaChi” and “Phi” at “Origin”, respectively. Perform a Omega scan with the condition above. Find the incident angle that gives the highest intensity. Input the value to “2Theta” and “Omega”.



☆In-Plane measurement (2ThetaChi/Phi scan)

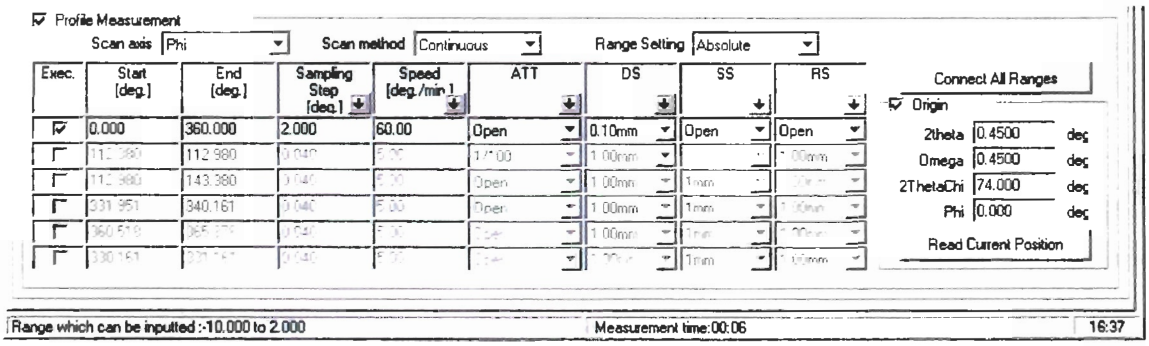
"2ThetaChi/Phi" is selected as "Scan axis". The scanning range is determined by referring to the ICDD card data. The sampling step should be 1/5 ~ 1/10 of a diffraction peak.

HINT : Any diffraction peaks can not be narrower than the resolution of the Soller slits (0.4deg. with standard Soller slit, and 0.12deg. with thin film PSA).



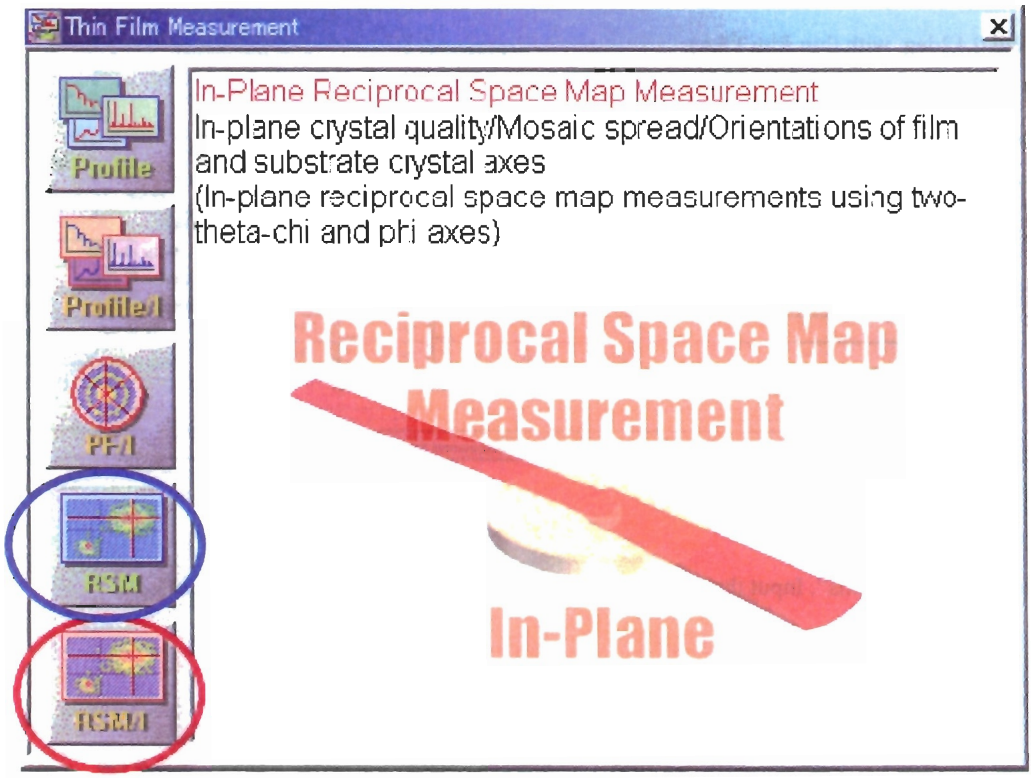
☆Rocking curve (Phi)

"Phi" is selected as "Scan axis". Input the 2Theta angle of the diffraction peak into "2ThetaChi".

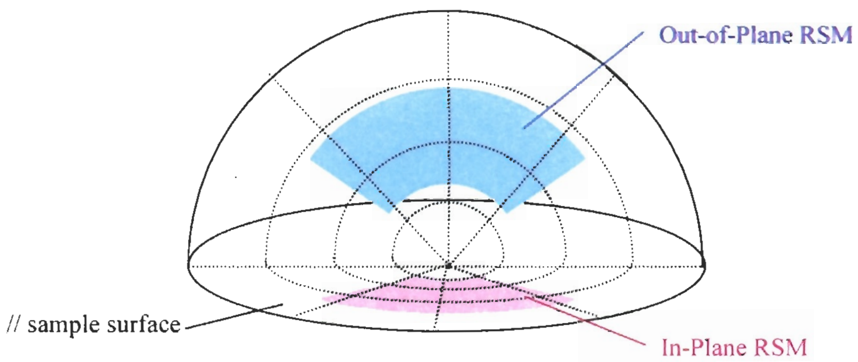


1-2-4 Reciprocal Lattice Map

Open [Start] – [Programs] – [Rigaku] – [Rigaku Measurement] – [Thin Film Meas. Menu].



Select [RSM] for Out-of-Plane reciprocal space mapping, or [RSM/I] for In-Plane reciprocal space mapping, respectively.



☆Specimen alignment

Perform the same procedures with Out-of-Plane or In-Plane measurements, respectively.

☆Preliminary measurement

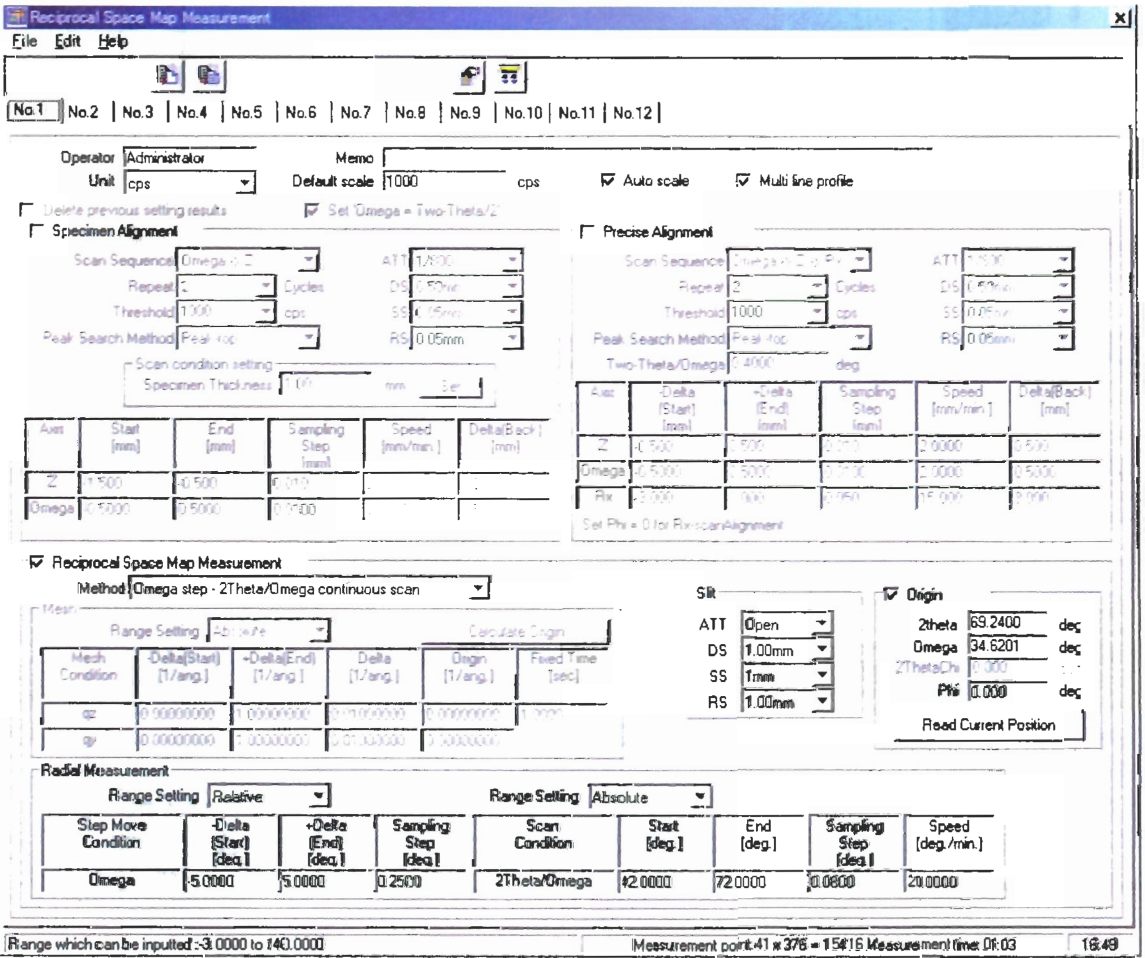
Refer to the results of 2Theta/Omeg (2ThetaChi/Phi) and Omega (Phi) scans to select the proper scanning ranges, sampling steps and scan speed.

☆Main measurement

A reciprocal space map is made of two dimensional data of d-value (calculated from 2Theta/Omega (2ThetaChi/Phi) scans) and crystal direction (obtained from Omega (Phi) scans). There are two ways to collect data;

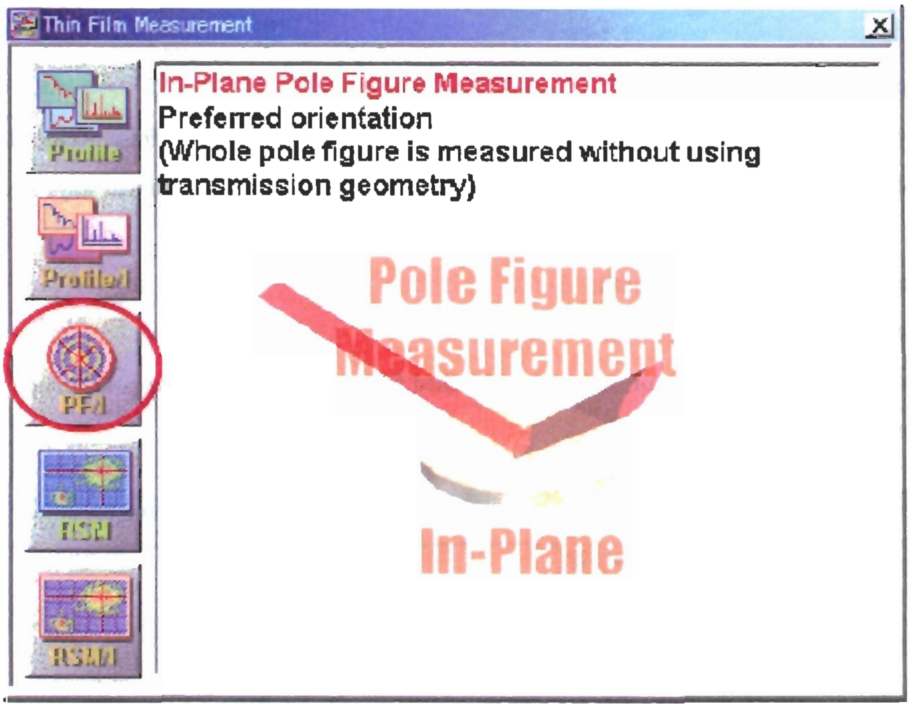
- Omega step – 2Theta/Omega scan
- 2Theta/Omega step - Omega scan

The mode which containes fewer number of scan will be finished faster than the other ; the positioning motion between each scan takes several minutes.

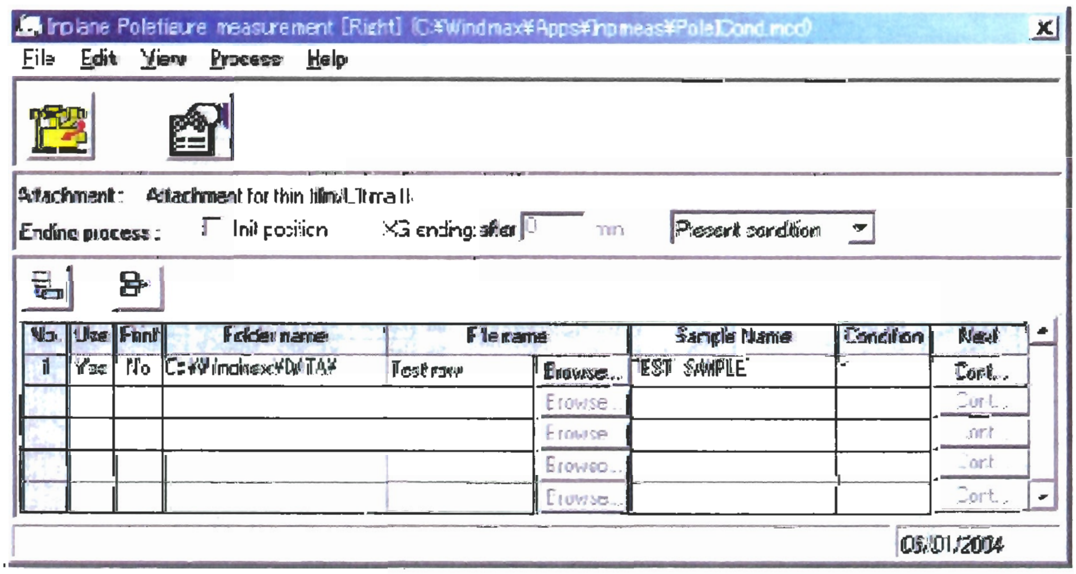


1-2-5 Pole Figure

Open [Start] – [Programs] – [Rigaku] – [Rigaku Measurement] – [Thin Film Meas. Menu].

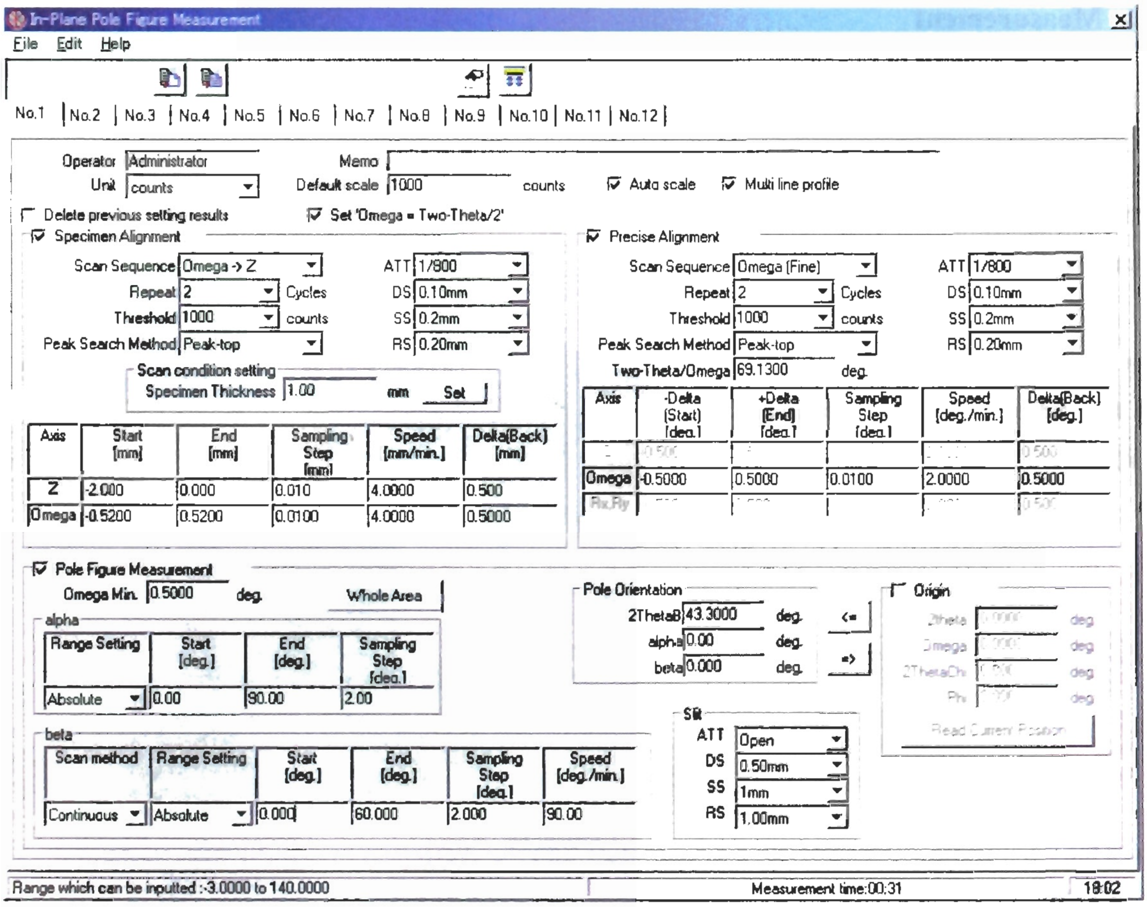


Select [PF/I] for pole figure measurement.



To edit scanning conditions, click on the "Condition edit" button or double click on the number input in [Condition].

The following is an example of specimen alignment / scanning conditions for pole figure menasutement



☆Specimen alignment

"Specimen alignment" has to be performed before pole figure measurement. Input "Specimen Thickness" and click "Set." The recommended scanning conditions of Z and Omega axes are input automatically.

"Precise Alignment" is used when users need to be sure the direction of lattice plane observed at the center of a pole figure (α=90deg.). When the critical angle for total reflection is input to "Two Theta/Omega," the lattice planes parallel to the sample surface are positioned at the center; when the diffraction angle of the substrate is input, the lattice planes parallel to the substrate's lattice planes are positioned at the center.

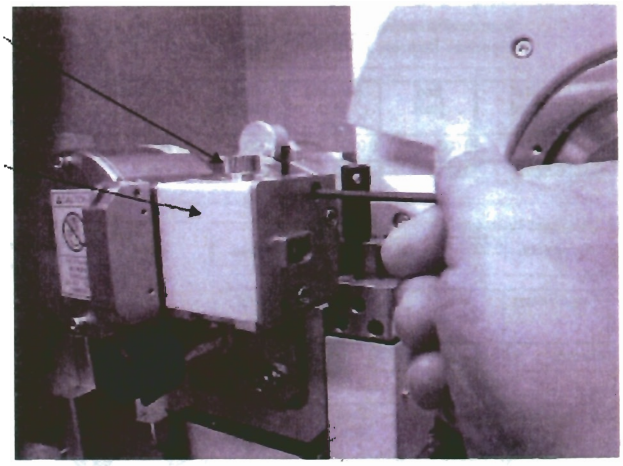
2. High- resolution x-ray diffraction

2-1 Measurement

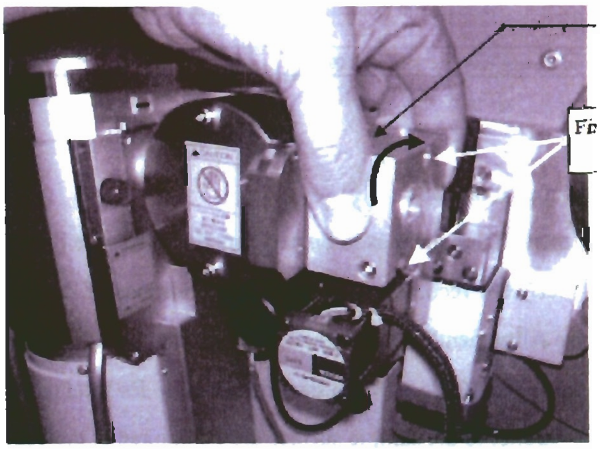
2-1-1 Setting up the geometry system

In this section, high- resolution optical system with Ge(220) incident monochromator is used to observe diffraction peaks from high-quality epitaxial films and single crystals. Adjust each optic by following the procedure mentioned in the manual.

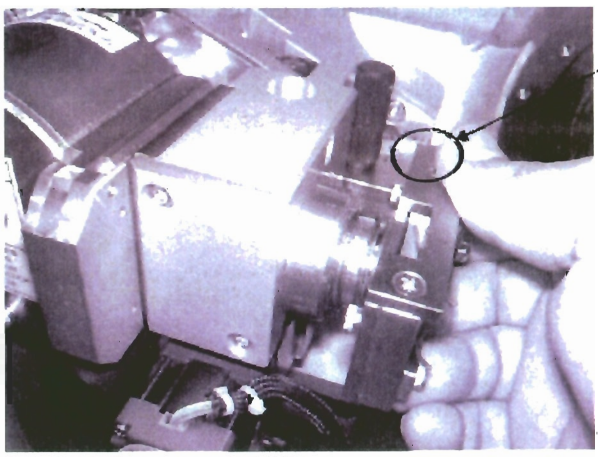
①Move θ-s to 45deg.
Remoe the incident-side slit and the incident Soller slit.



Attaching the incident Ge(220) channel cut Monochromator.



Attaching the dedicated incident- side slit.



TTRAXIII

Open “Rigaku control panel” from the Rigaku folder. Double click “RINT 2200 Rigaku system. ”

Select following items;

Attachment:

Attachment for thin film/

UltimaIII

Incident monochro:

Use

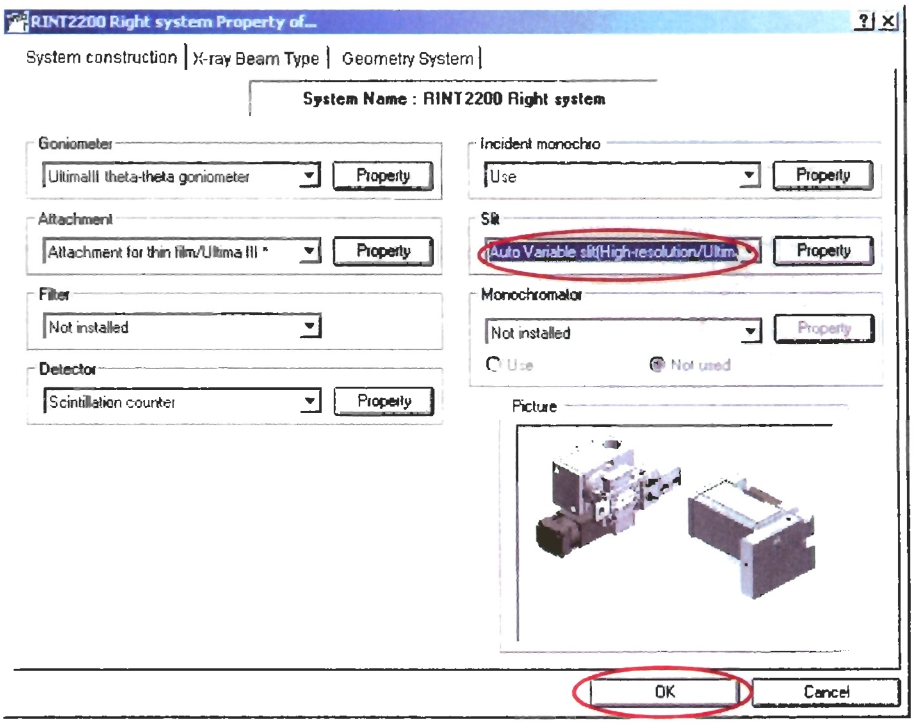
Slit:

Auto variable slit

(High- resolution/UltimaIII)

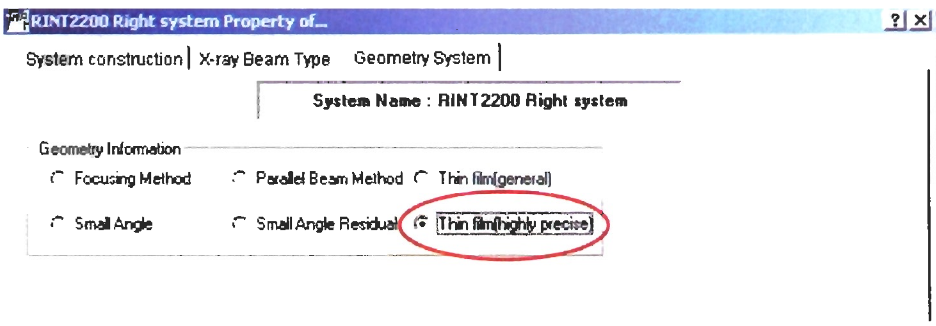
Monochromator:

Not installed



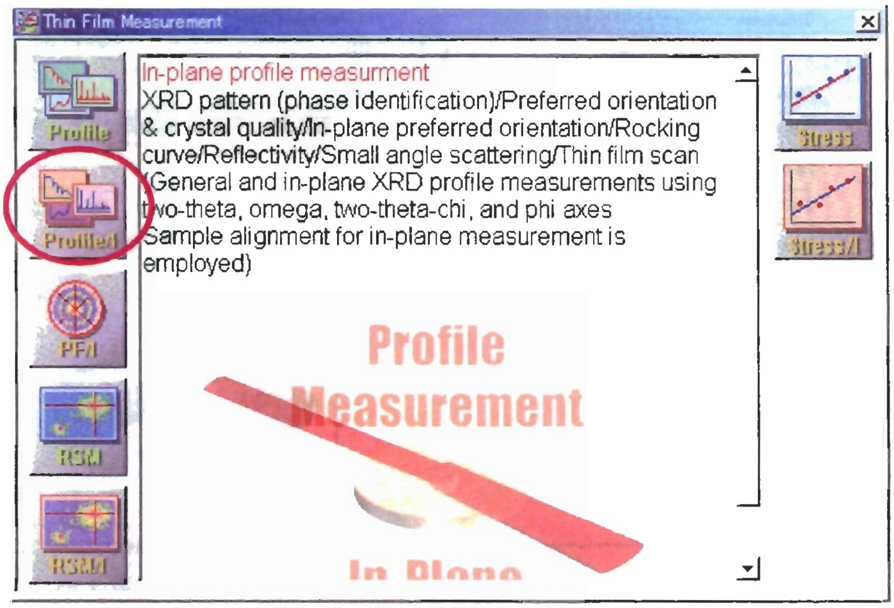
Geometry Information:

Thin film (highly precise)

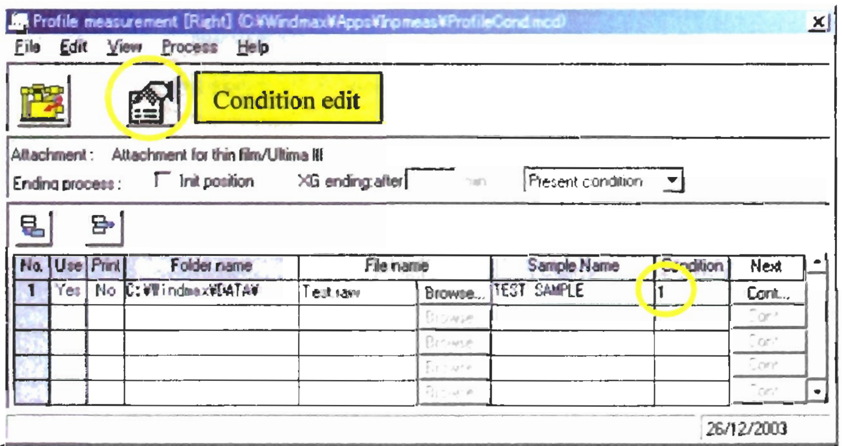


2-1-2 Rocking curve Measurement

Open [Start] – [Programs] - [Rigaku] – [Rigaku Measurement] – [Thin Film Meas. Menu.]



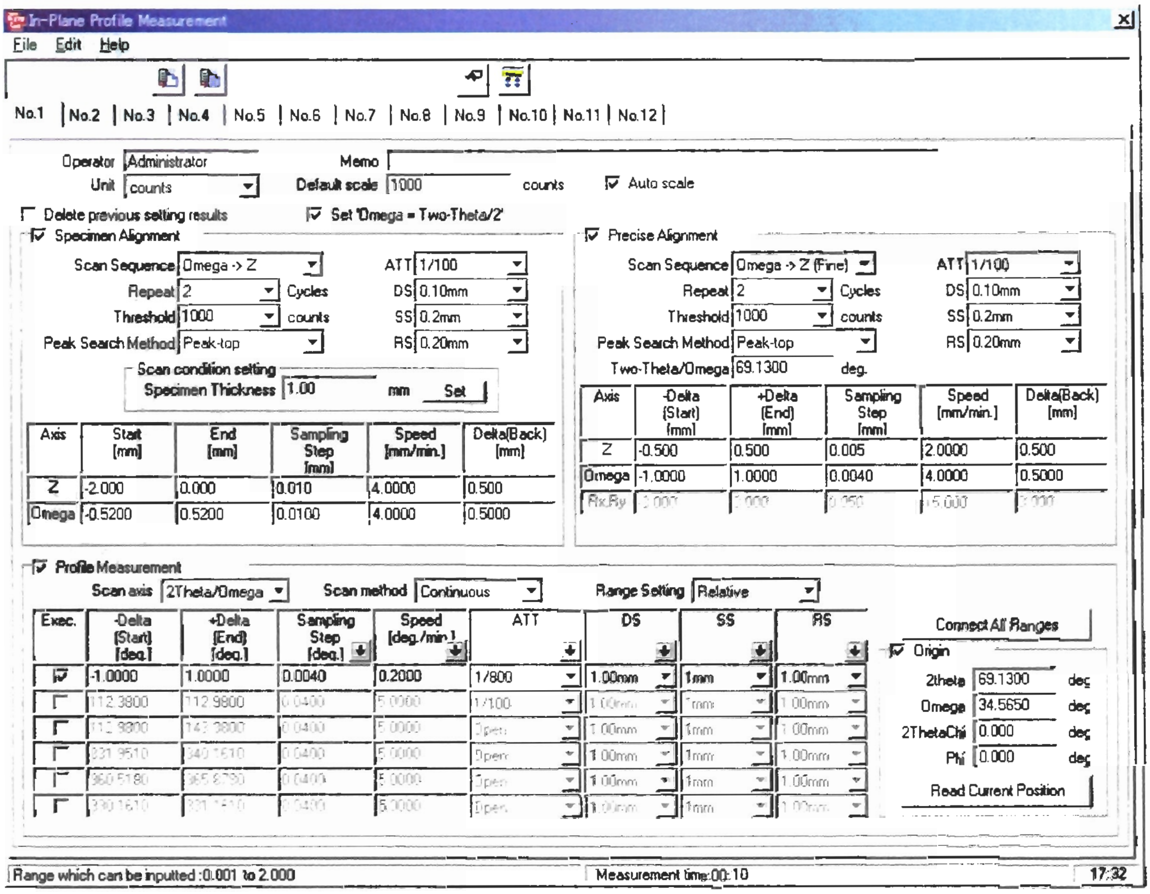
Select [Profile/In-Plane] measurement.



To edit scanning conditions, click on the "condition edit" button or double-click on the number input in [Condition.]

☆Specimen alignment

XRD measurement for epitaxial films and single crystal substrates requres precise azimuth alignment because the distribution width of reciprocal lattice points are narrow sot hat the each goniometer axis should be arranged precisely.



☆High resolution rocking curve (symmetric scan)

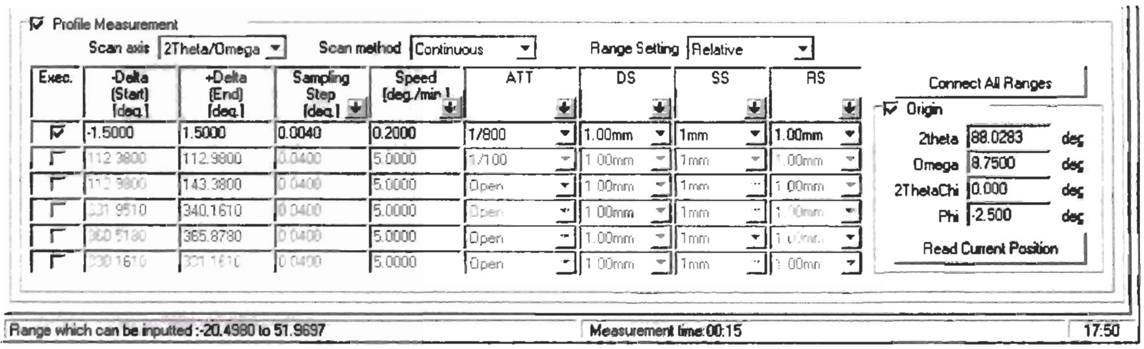
The symmetric rocking curve observes the crystal planes almost parallel to the sample surface. Since the "Precise Alignment" procedure makes those crystal planes being observed, perform 2theta.omega scan right after the alignment. Input the diffraction angle of the substrate into "2Theta" and the half value into "Omega" in "Origin", respectively.

☆High- resolution rocking curve (asymmetric scan)

To observe crystal planes not parallel to the sample surface, the difference between incident and exit angles should be equal to the angle between the observing crystal planes and the symmetric planes. The angle between $(h_1k_1l_1)$ and $(h_2k_2l_2)$ is calculated with the following equation (for cubic system),

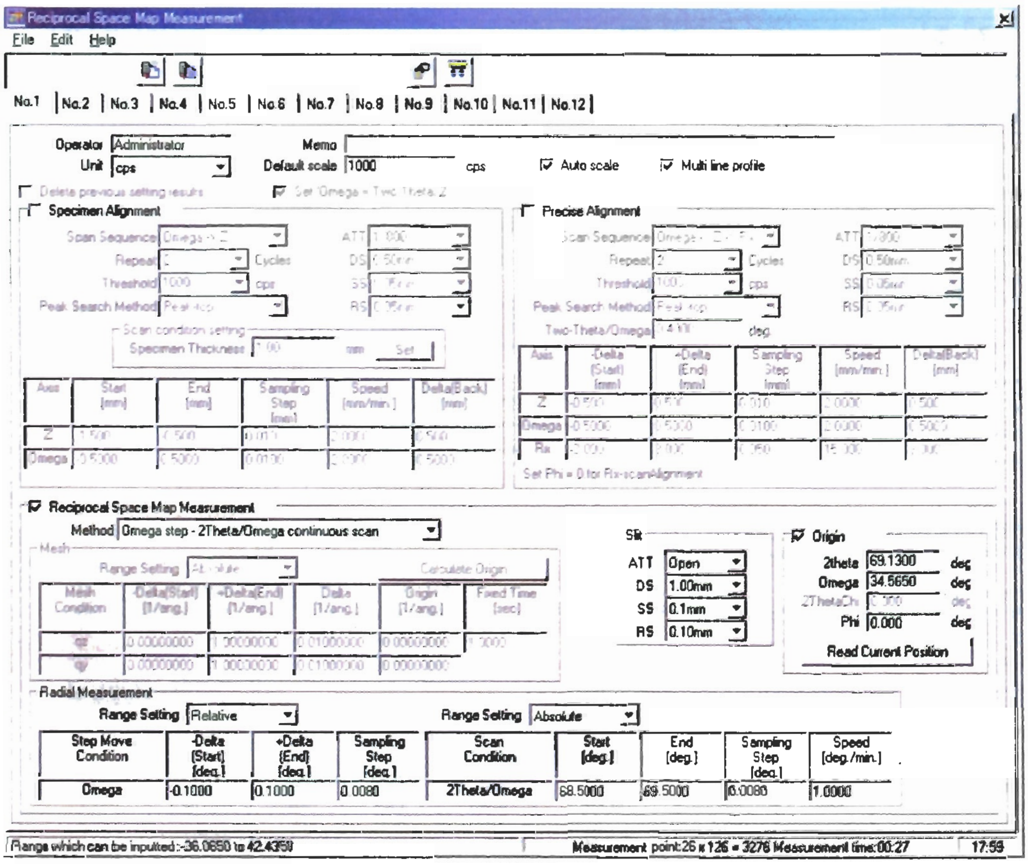
$$\text{Cubic system :}\quad \cos\phi = \frac{h_1h_2 + k_1k_2 + l_1l_2}{\sqrt{\left(h_1^2 + k_1^2 + l_1^2\right)\left(h_2^2 + k_2^2 + l_2^2\right)}}$$

Also phi angle (in-plane sample rotation) should be at proper position to observe asymmetric planes. Perform phi scan before starting main measurements and find the proper position.



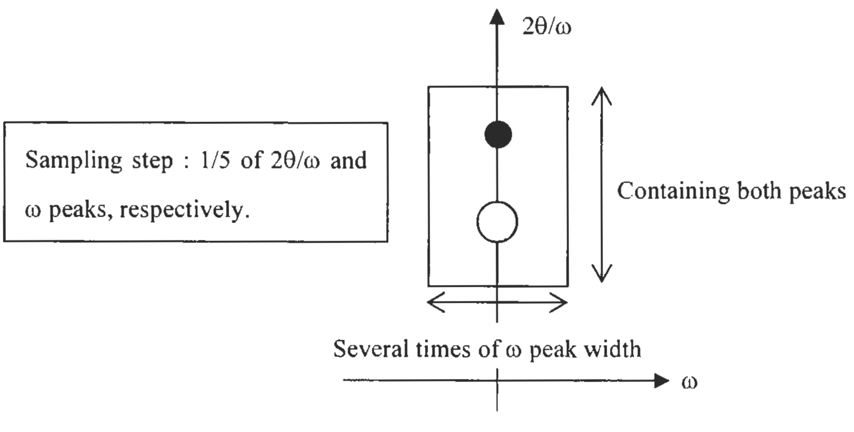
2-1-3 Reciprocal Lattice Mapping

Reciprocal lattice mapping is used to obtain the coordinates of reciprocal lattice points and the degree of relaxation for epitaxial films. An example of scanning condition for SiGe (004) / Si (004) samples are as follows;



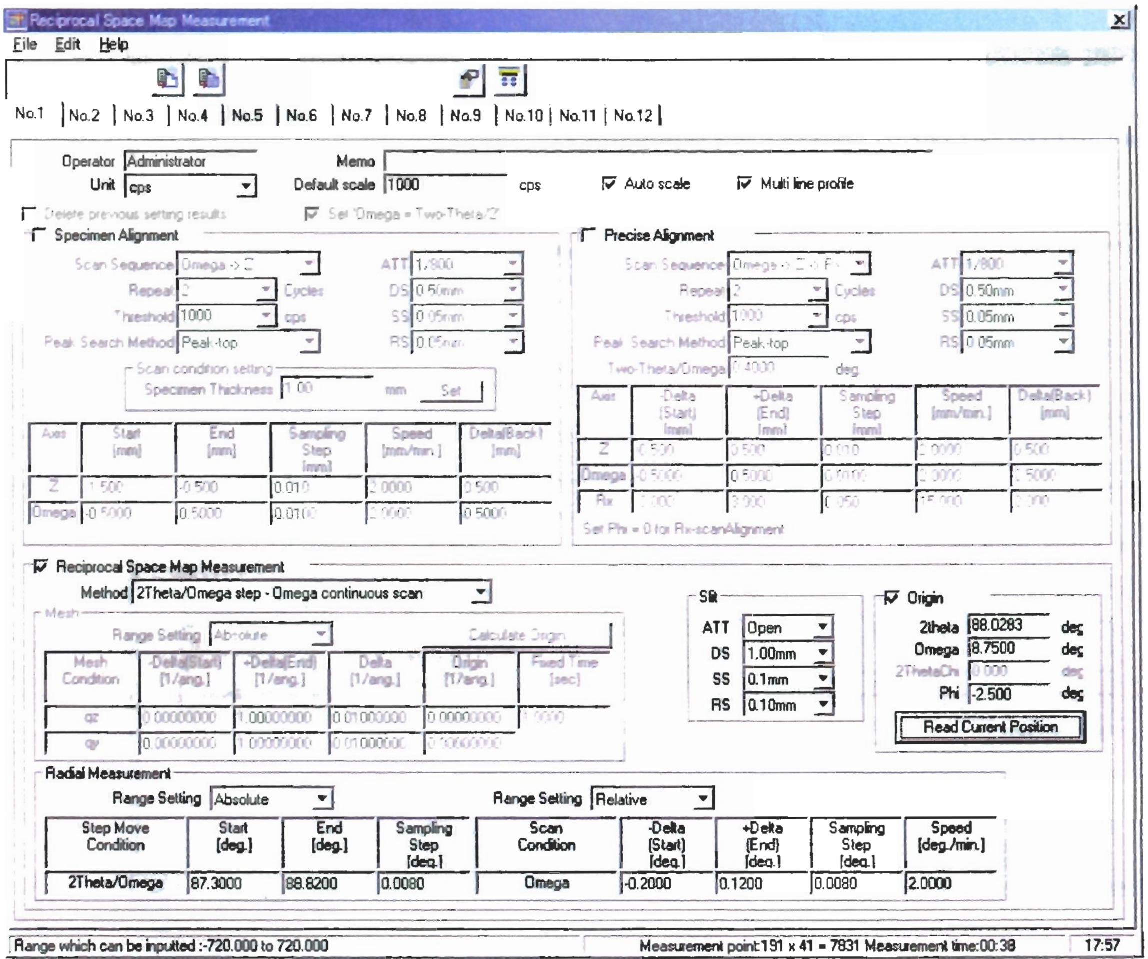
Measurement mode (Omega step – 2Theta/Omega scan or 2Theta/Omega step – Omega scan) is selected as the total number of scan becomes fewer. Because reciprocal space map consists of successive 1D (2θ/ω or ω) scans and it takes several ten seconds to position scan axes (axis) to the start angle in every intervals.

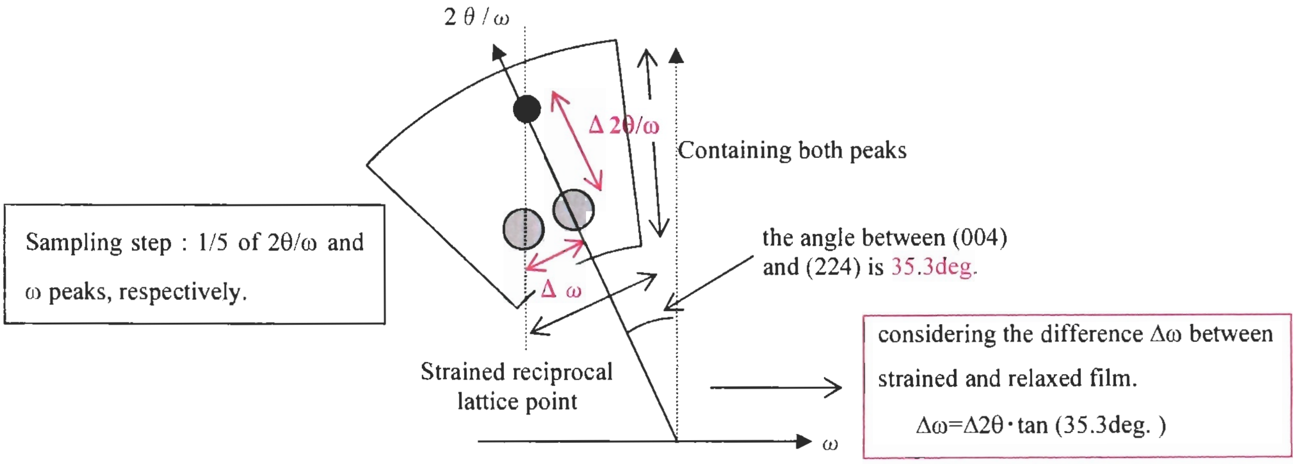
To observe the spread of reciprocal lattice point, the range of measurement should be wide enough. The scan range of 2θ/ω is determined by referring to rocking curve profile as the intensity decreases to the background level at the both end. The range of Omega should be several times larger than the ω peak width.



The following is a example of the scanning condition for SiGe (224) / Si (224) samples.

Considering the epitaxial films can be totally strained, the range of Omega should be wider than the case of symmetric RSM measurement.

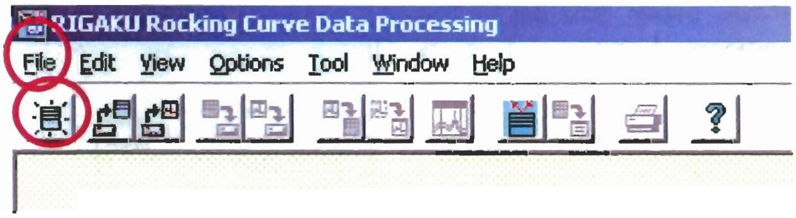




2-2 Outline of rocking curve simulation software

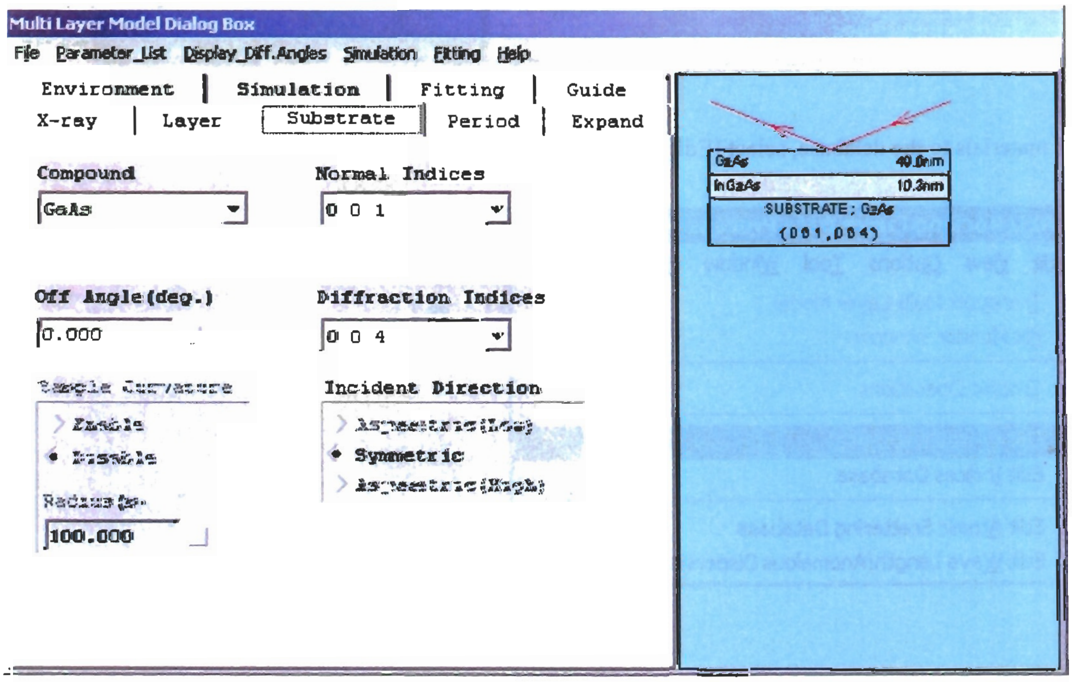
2-2-1 Sample structure and x-ray divergence

Click  or select [File] - [New].

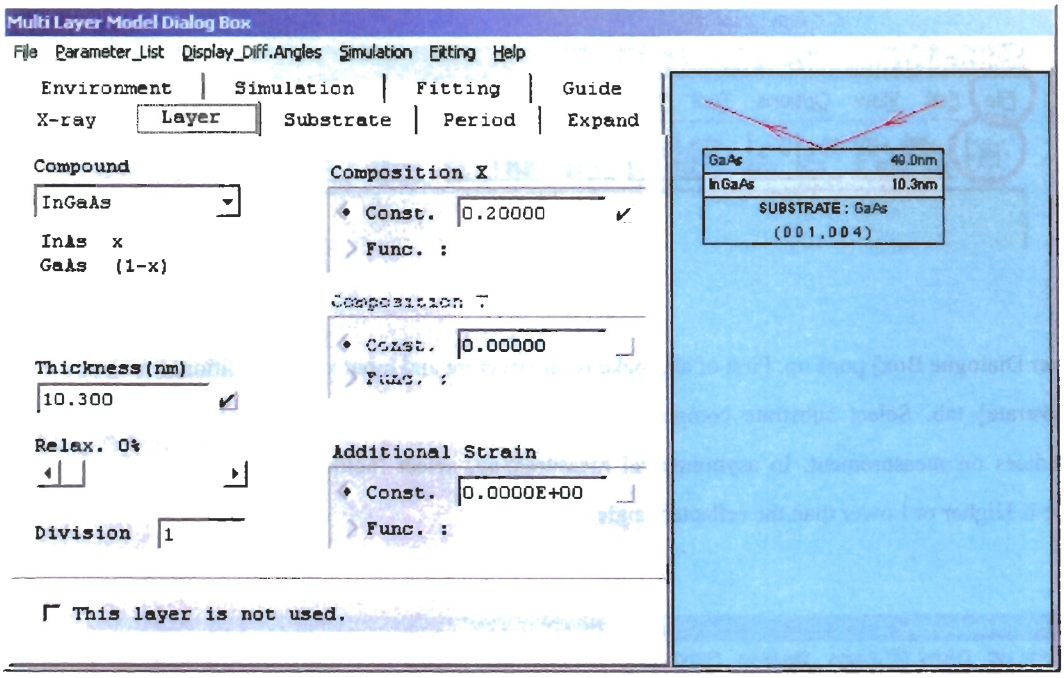


[Multi Layer Dialogue Box] pops up. First of all, make layer structure and input x-ray condition.

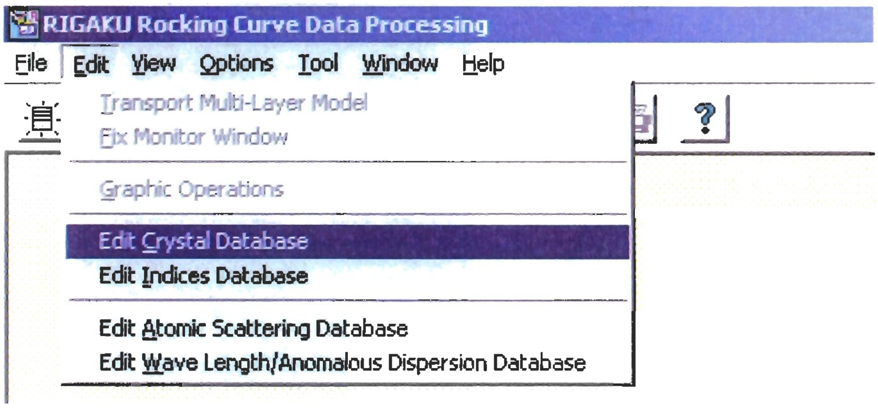
Click [Substrate] tab. Select substrate compound, surface normal indices, offset angle if it has, and diffraction indices on measurement. In asymmetrical measurement, select Incident Direction based on the incident angle is Higher or Lower than the reflected angle.



To add films, double- click SUBSTRATE in the illustration. Select compound, input layer thickness and percentage of the composition X in the case of solid solution. Layers are copied/ deleted by double- clicking on the film in the illustration with left/ right mouse button, respectively.

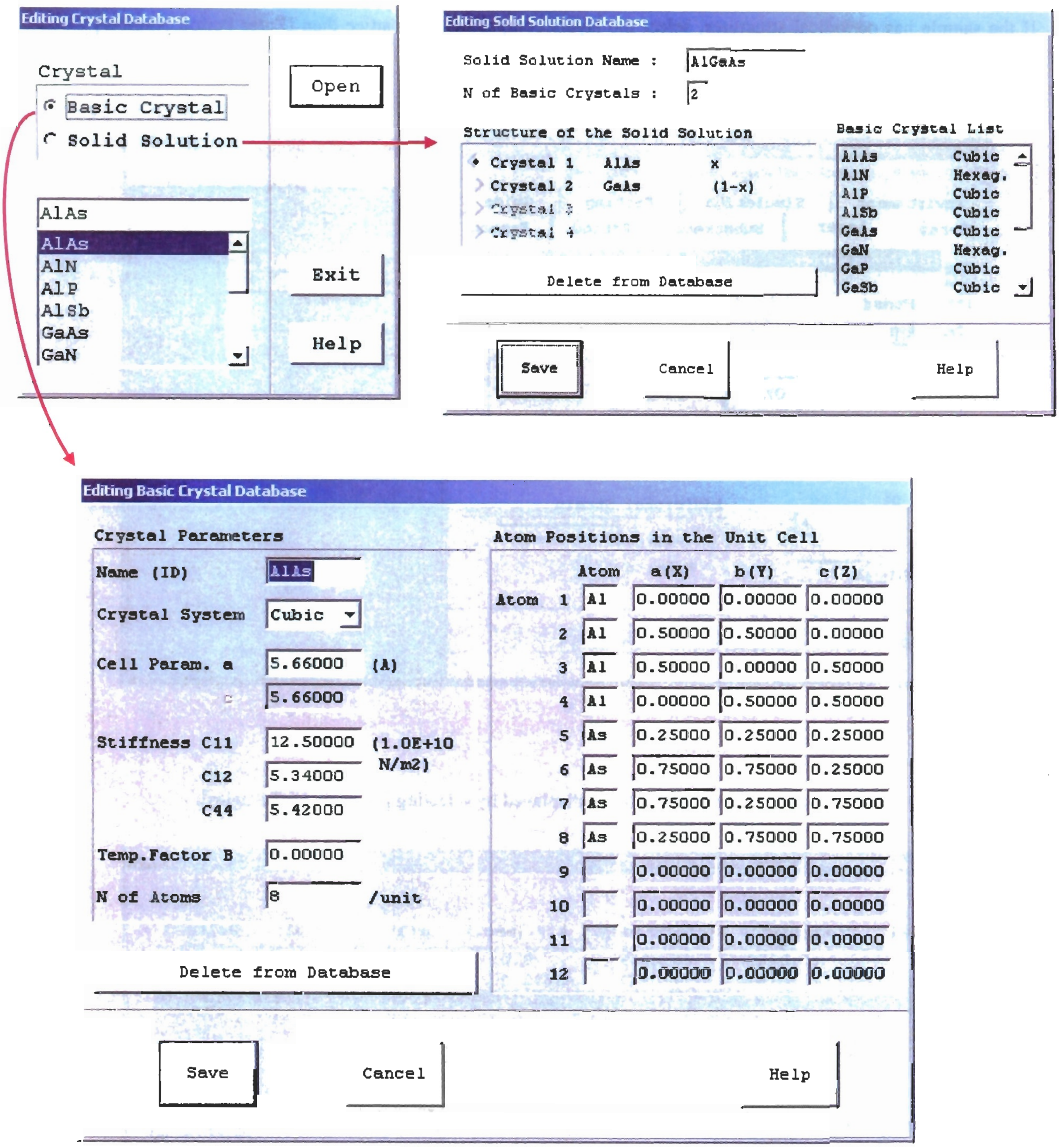


To add new materials to the database, select [Edit] - [Edit Crystal Database] .

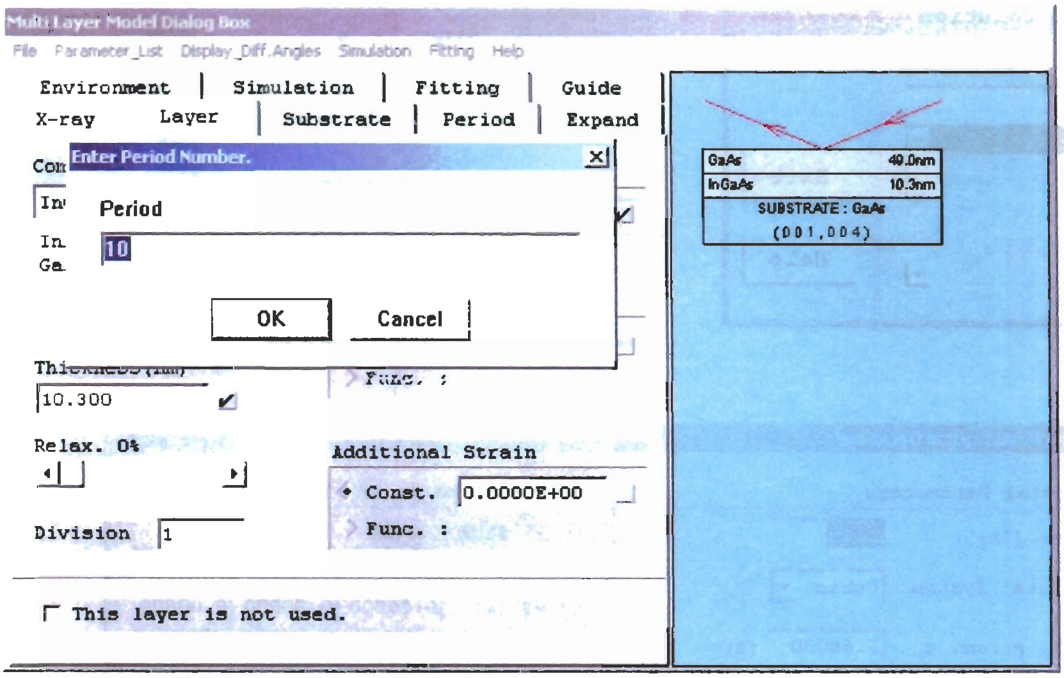


Elements (e.g. Si and Ge) and binary compounds (e.g. GaAs) are included in Basic Crystal list. Crystallographical coefficients are necessary to add new Basic Crystal to the database.

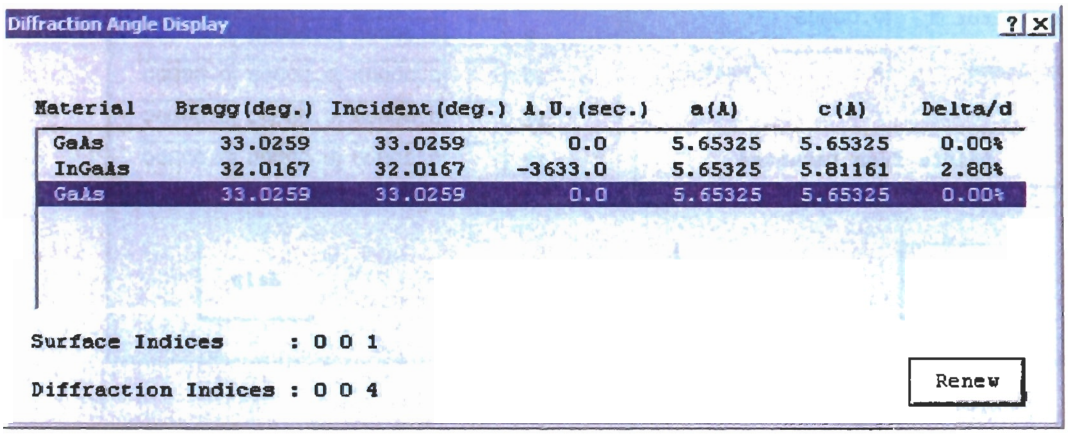
Solid solution consists of two basic crystals. New solid solution model is registered if the basic crystals have common crystal system. The values of crystallographical coefficients become the linear combination of each basic crystal.


Editing Crystal Database
Crystal
Basic Crystal
Solid Solution
AlAs
AlAs
AlN
AlP
AlSb
GaAs
GaN
Open
Exit
Help
Editing Solid Solution Database
Solid Solution Name :
AlGaAs
N of Basic Crystals :
2
Structure of the Solid Solution
Crystal 1 AlAs x
Crystal 2 GaAs (1-x)
Crystal 3
Crystal 4
Delete from Database
Basic Crystal List
AlAs Cubic
AlN Hexag.
AlP Cubic
AlSb Cubic
GaAs Cubic
GaN Hexag.
GaP Cubic
GaSb Cubic
Save
Cancel
Help
Editing Basic Crystal Database
Crystal Parameters
Name (ID) AlAs
Crystal System Cubic
Cell Param. a 5.66000 (A)
c 5.66000
Stiffness C11 12.50000 (1.0E+10 N/m2)
C12 5.34000
C44 5.42000
Temp.Factor B 0.00000
N of Atoms 8 /unit
Delete from Database
Atom Positions in the Unit Cell
Atom a(X) b(Y) c(Z)
Atom 1 Al 0.00000 0.00000 0.00000
2 Al 0.50000 0.50000 0.00000
3 Al 0.50000 0.00000 0.50000
4 Al 0.00000 0.50000 0.50000
5 As 0.25000 0.25000 0.25000
6 As 0.75000 0.75000 0.25000
7 As 0.75000 0.25000 0.75000
8 As 0.25000 0.75000 0.75000
9 0.00000 0.00000 0.00000
10 0.00000 0.00000 0.00000
11 0.00000 0.00000 0.00000
12 0.00000 0.00000 0.00000
Save
Cancel
Help

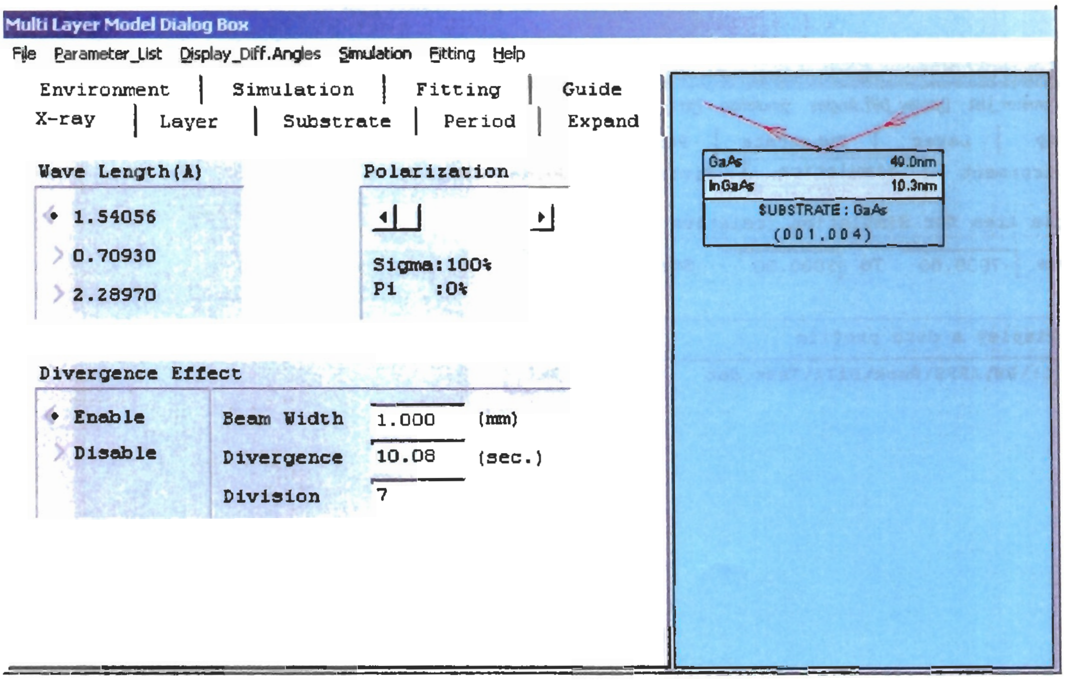
If the sample has periodical structures, select the layers composing superlattice then [Enter Period Number] window pops up. Input the number of time.



The diffraction angle of those films and substrate are displayed by selecting [Display_Diff.Angles].

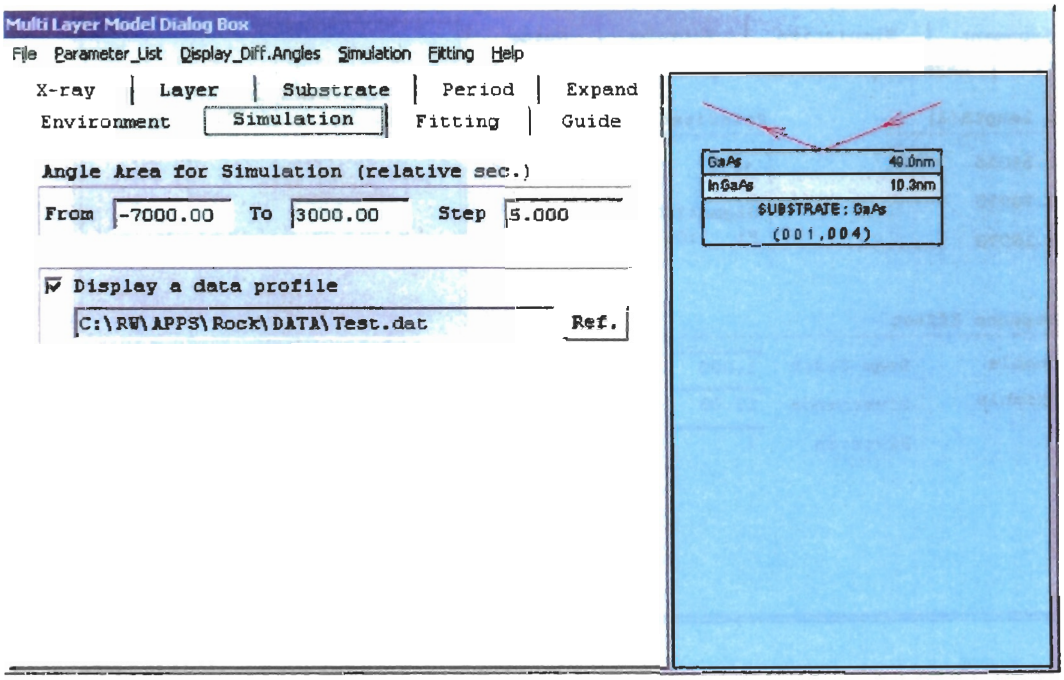


Input x-ray wavelength, polarization and divergence. The divergence angle is selected by incident optical system. For [Polarization], σ polarization should normally be selected.

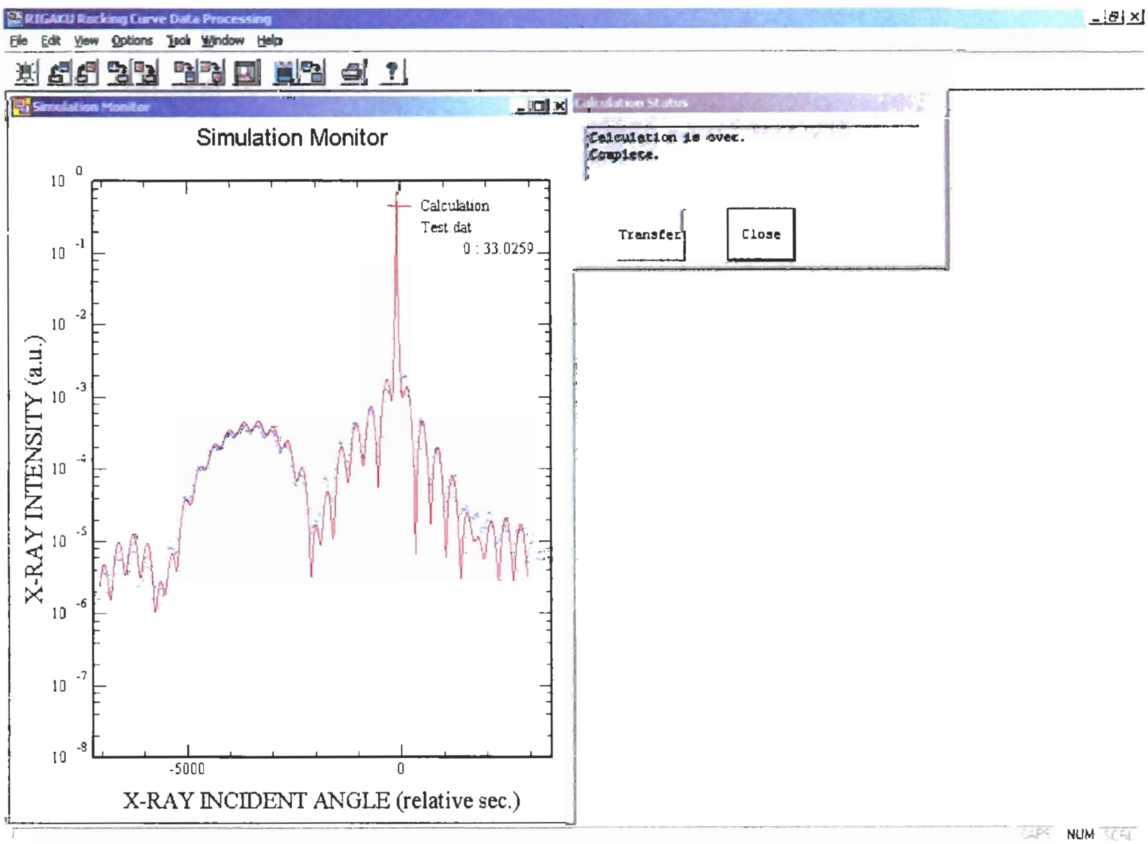


2-2-2 Comparison with measured profile

To compare calculated profile to experimental data, click [Simulation] tab, check [Display a data profile] and select experimental file. Input simulation area and step then click [Simulation] on the bar above.

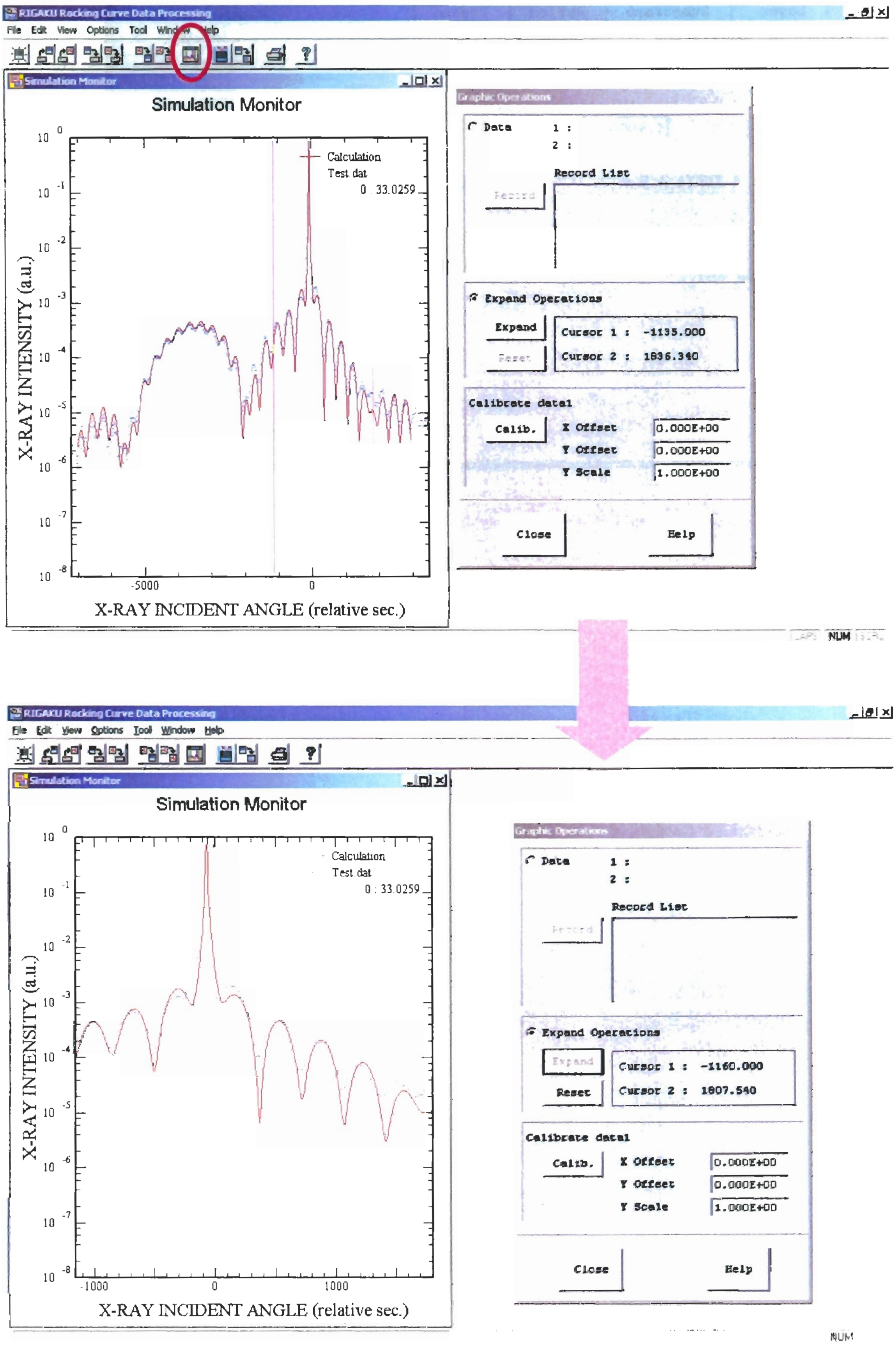


Calculated profile is shown in [Simulation Monitor] .

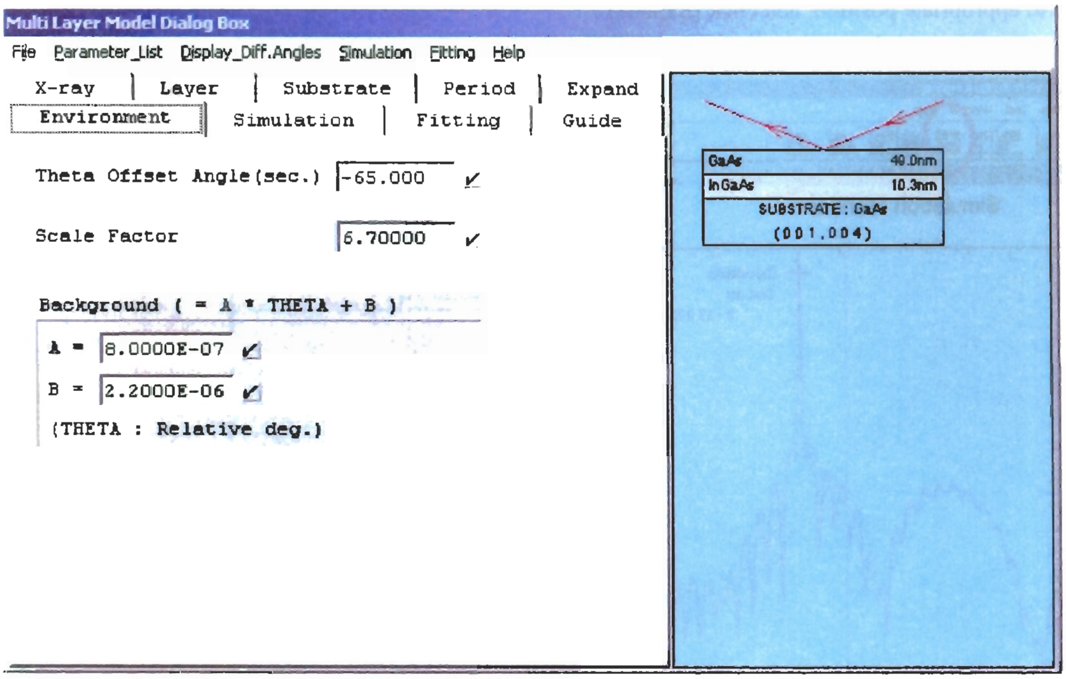


2-2-3 Expanding

To expand a part of a profile, click [icon] or select [Edit] – [Graphic Operation] . Check [Expand Operations], move cursors to appropriate position, and click [Expand] .

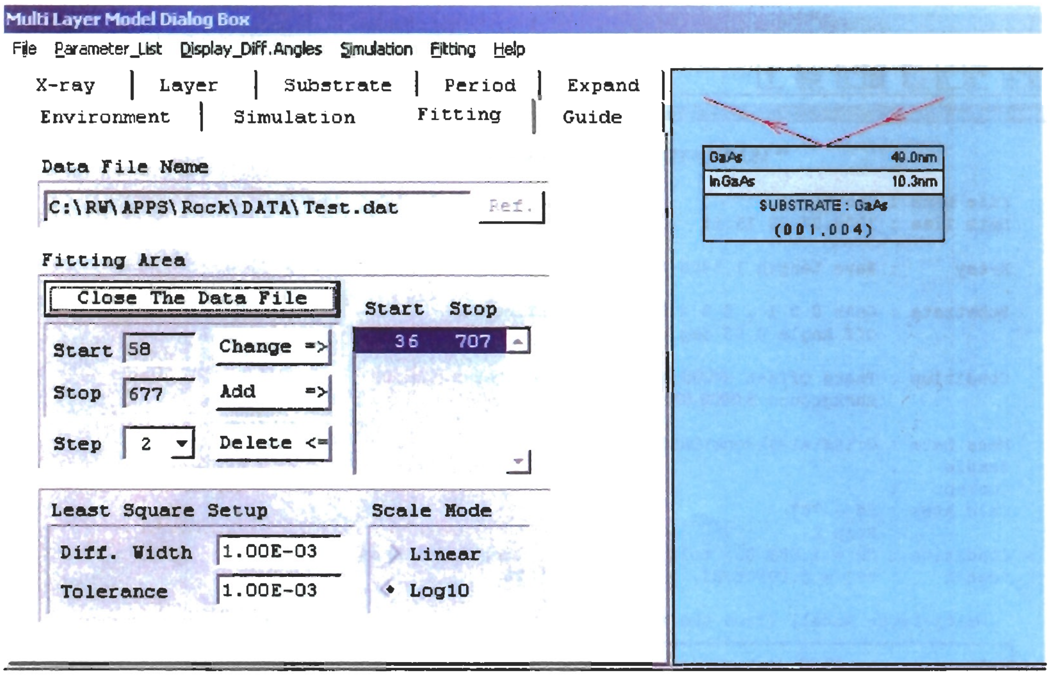


If the peak angle differs from each other, make an offset on the calculated profile. Click [Environment] tab and input offset value. Also dynamic scale and background level are entered in this tab.

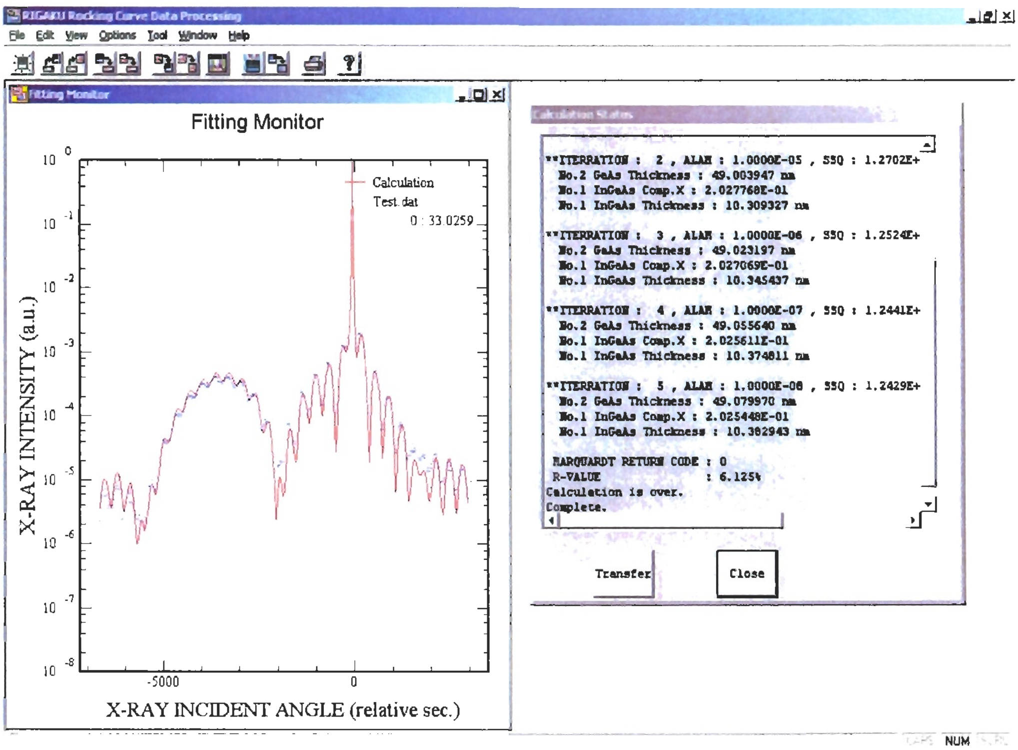


2-2-4 Profile Fitting

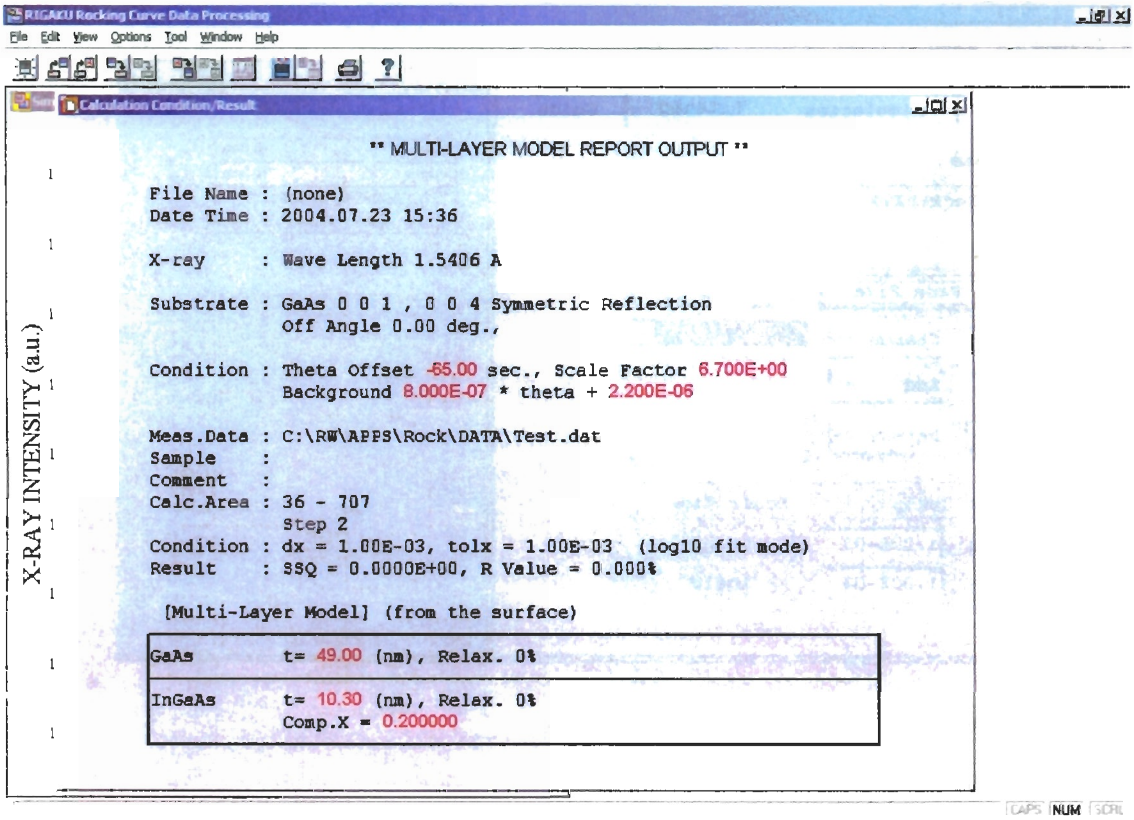
Final procedure of rocking curve analysis is profile fitting. Click [Fitting] tab, select measurement data, and input fitting area. Parameter which have checkmark in their checkbox are refined.



To start fitting procedure, click [Fitting].



The result of fitting is shown by selecting [Parameter_List] on [Multi Layer Model Dialog Box]. When clicking the menu, a message asks whether you want to display the fitting conditions. By choosing "Yes", fitted parameters are displayed by red characters.



3. X-ray reflectivity

3-1 Flow Chart

Preparation

To select the geometry system, sampling step and slit width, simulate XRR profile with GXRR software. Click and input designed layer structure.

☞ To select optical system, change [Number of divisions] from 1 to 7. Input 0.04 and 0.01 into [Divergence] and examine the profiles;

If the profiles do not change clearly →Slit collimation

If 0.01 gives distinct oscillation while 0.04 gives dull shape

→Ge(220)channel-cut monochrometer

☞ To select step width, change [step] value and compare the profiles.

☞ To select the DS slit, estimate the critical angle. To observe XRR profiles properly, it is necessary to limit the irradiation width within the sample width at critical angle. Refer to [irradiation width- incident angle] graph and select the most suitable one.

Measurement

☞ Optics adjustment.

☞ Sample alignment and measurement. Perform 2theta/omega scan with the sampling step selected above.

Analysis

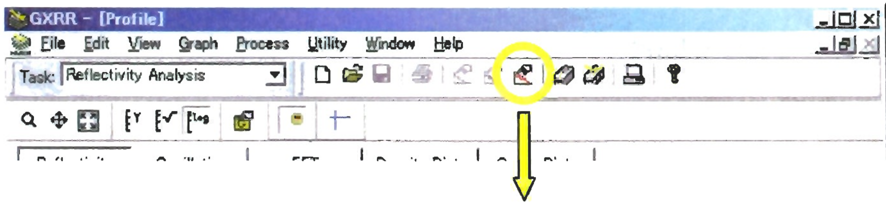
☞ RMS fitting with GXRR software. Click to switch to fitting mode. Compare calculated profile to measured profile, change parameters to make it resemble measured one before execute profile fitting.

☞ If there are large differences between calculated and measured profile, the designed layer structure might be quite different from the actual structure. In such a case, Fourier Transformation would be effective to make a proper layer model.

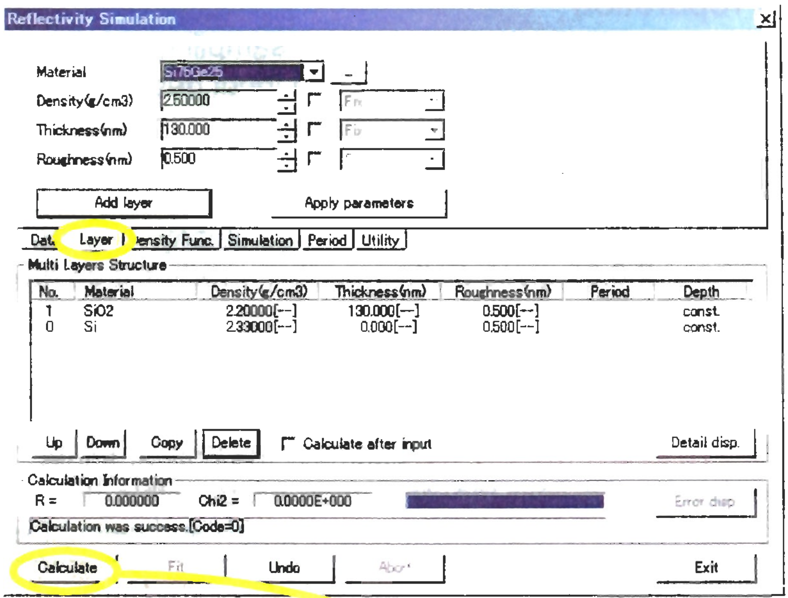
3-2 Preparation

Start GXRR software.

Click “Simulation” button.



Select “layer” tab.



Input the designed layer structure.

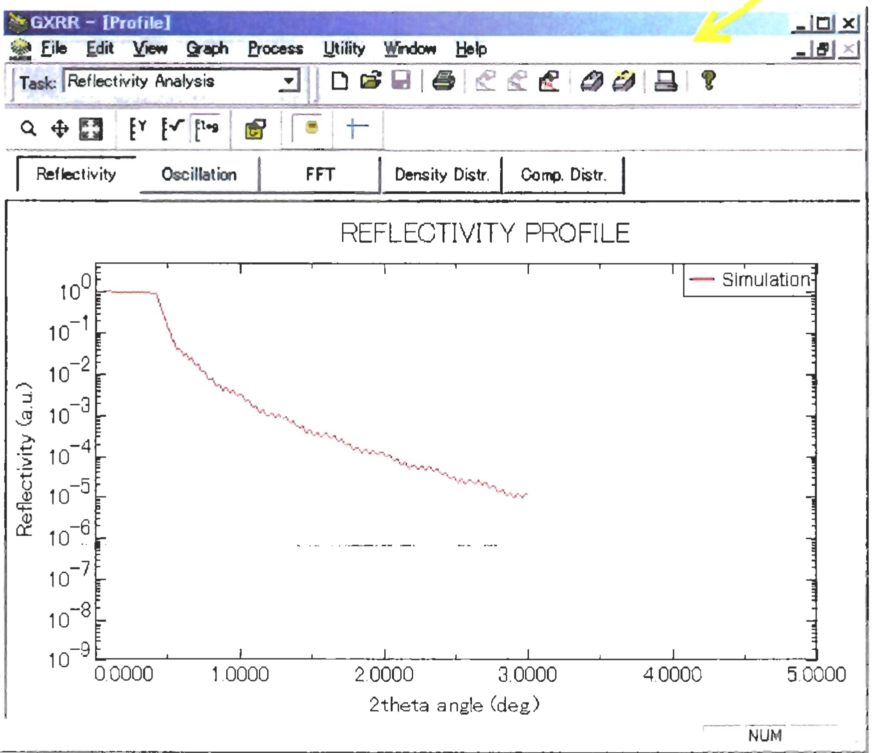
Choose the substrate/layer material from “Material” list.

Click “Add layer” button.

Change “Thickness” to the designed values.

Use “Up” and “Down” buttons to switch the order of the layers.

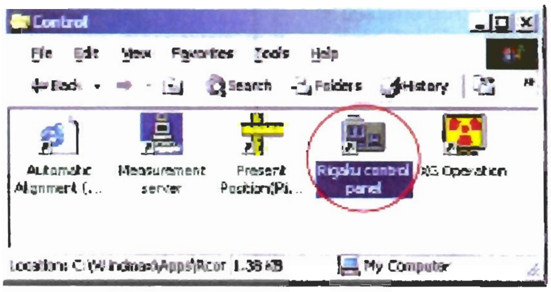
Click “Calculate” to show the simulated profile. (When “Calculate after input” is checked, the simulated profile will be calculated automatically.)

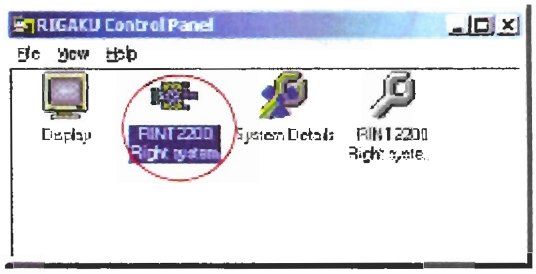


3-3 Measurement

3-3-1 Setting up the geometry system

Open "Rigaku control panel" from the Rigaku folder. Double click "RINT 2200 Rigaku system. "





Select following items;

Attachment :

Attachment for thin film/ UltimaIII

Incident monochro:

Use

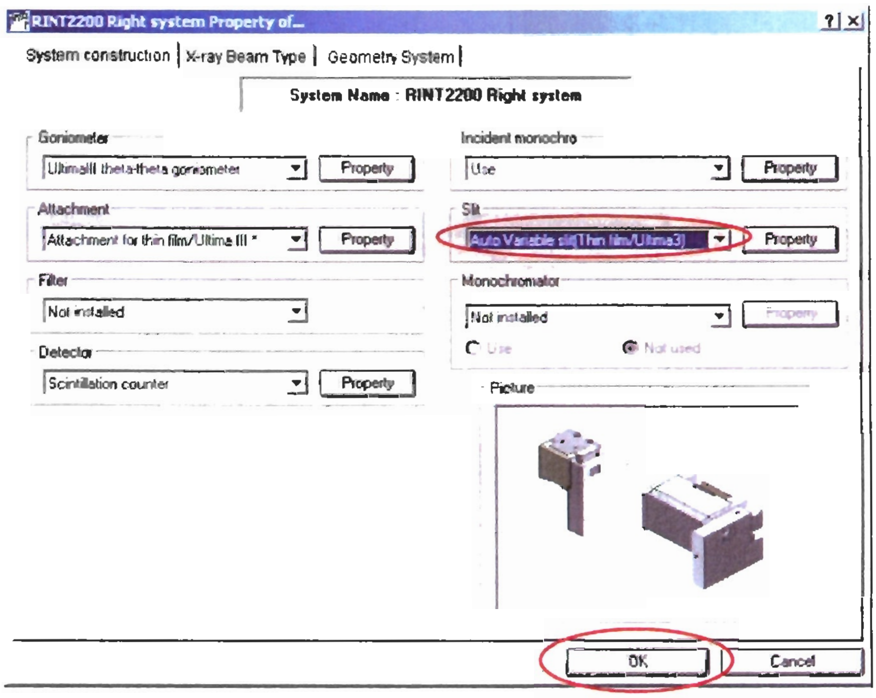
Slit:

Auto Valiable slit(Thin film/

UltimaIII)

Monochrometer:

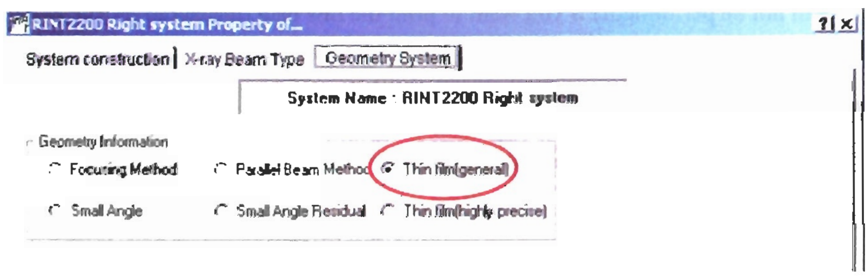
Not installed



Geometry Information:

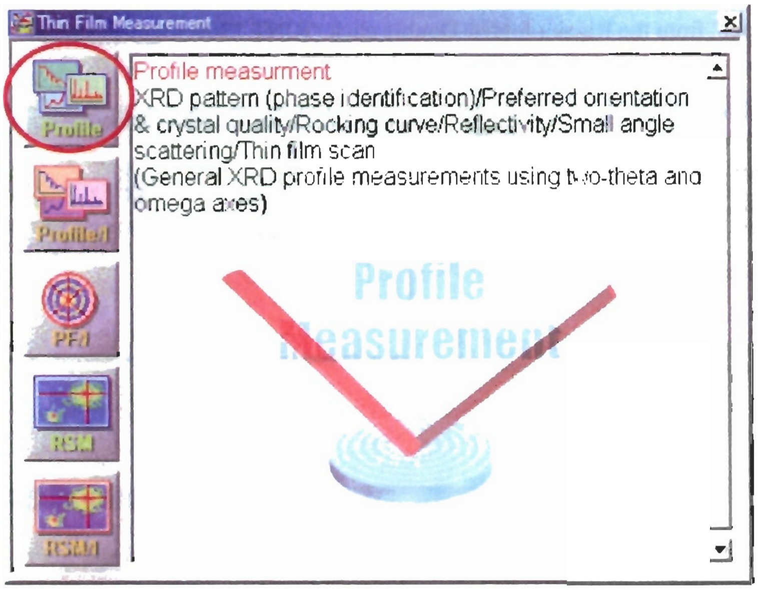
Thin film (general) or

Thin film (highly precise)

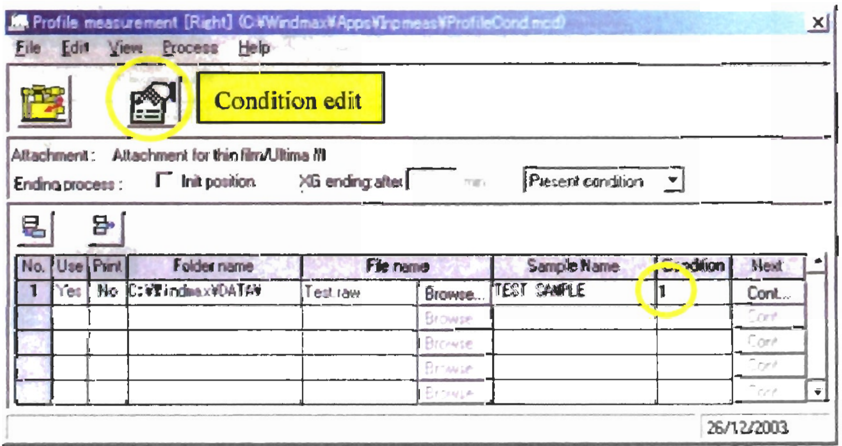


3-3-2 XRR Measurement

Open [Start] – [Programs] - [Rigaku] – [Rigaku Measurement] – [Thin Film Meas. Menu.]

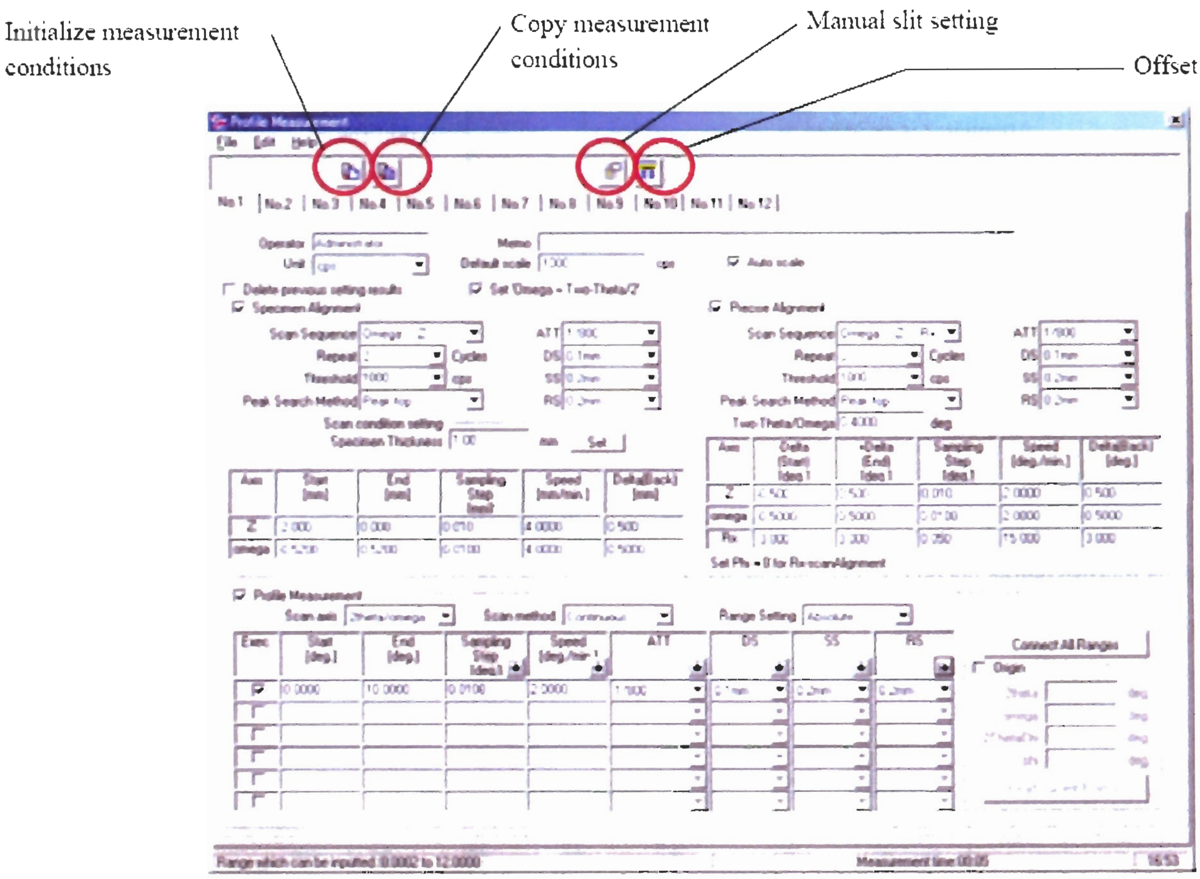


Select [Profile] measurement.



Input the folder name, the file name and the sample name. Switch "Use" to "Yes" (otherwise the measurement will not be performed!) If you want to print the raw data (measured profile and the scanning condition) after the measurement, switch "Print" to "Yes."

To edit measurement conditions, click on the "condition edit" button or double-click on the number input in [Condition.]



☆Specimen alignment

For X-ray reflectivity analysis, both "Specimen Alignment" and "Precise Alignment" are needed to be done before the measurement. Check these items.

"Specimen Alignment" – Input "Specimen Thickness" and click "Set. " The recommended scanning conditions of Z and omega axes are input automatically.

"Precise Alignment" – Input the value little bit smaller than the critical angle to "Two Theta/Omega. "

☆Preliminary measurement

To estimate the attenuator switching positions, preliminary measurement is performed. The following is the typical scanning condition.

Two theta / omega scan range : 0-3deg.

Sampling step : the estimated value from the simulation

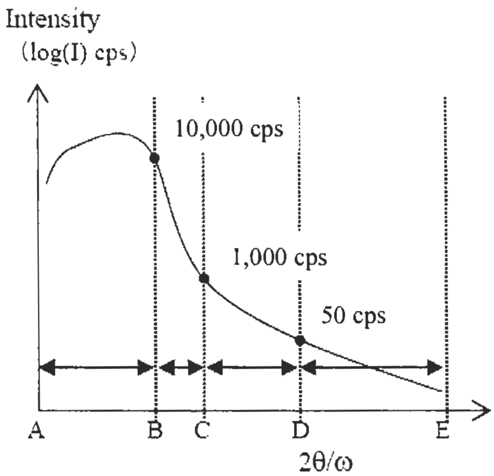
Sampling speed : 1deg./min.

Attenuator : 1/800

DS, SS, RS : the selected widths from the simulation.

The attenuator switching positions are estimated as follows.

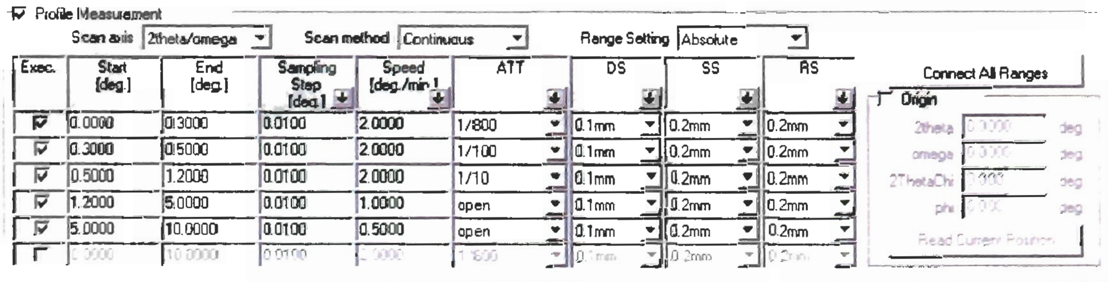
Intensity	Attenuator is switched as
10,000 cps	1/5000 → 1/400
1,000 cps	1/400 → 1/30
50 cps	1/30 → open



☆Main measurement

Remove the check marks at "Specimen Alignment" and "Precise Alignment" because the alignment procedures are already finished before the preliminary measurement.

Set the scan conditions and attenuator switching positions for main measurement. Click on "Connect All Ranges" to make sure that the start and end angle of each scan is continuous.



Profile Measurement

Scan axis: 2theta/omega   Scan method: Continuous   Range Setting: Absolute

Exec.	Start [deg.]	End [deg.]	Sampling Step [deg.]	Speed [deg./min.]	ATT	DS	SS	RS
☑	0.0000	0.3000	0.0100	2.0000	1/800	0.1mm	0.2mm	0.2mm
☑	0.3000	0.5000	0.0100	2.0000	1/100	0.1mm	0.2mm	0.2mm
☑	0.5000	1.2000	0.0100	2.0000	1/10	0.1mm	0.2mm	0.2mm
☑	1.2000	5.0000	0.0100	1.0000	open	0.1mm	0.2mm	0.2mm
☑	5.0000	10.0000	0.0100	0.5000	open	0.1mm	0.2mm	0.2mm
☐	[illegible]	10.0000	0.0100	[illegible]	[illegible]	[illegible]	0.2mm	[illegible]

Connect All Ranges

Origin: 2theta, omega, 2ThetaChi, phi

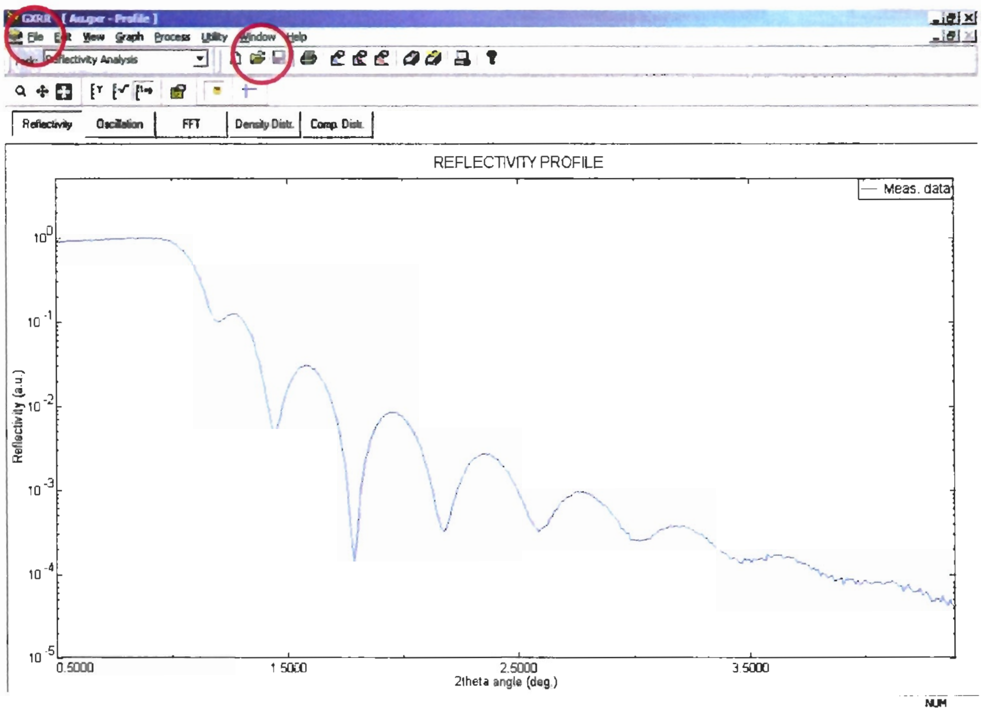
3-4 Outline of XRR analysis software (Demo data file path : C:¥Program Files¥Rigaku¥GXRR3¥Data)

3-4-1 Open experimental data

Start the GXRR software and read experimental data. Both of *.raw and *.dat are readable on GXRR.

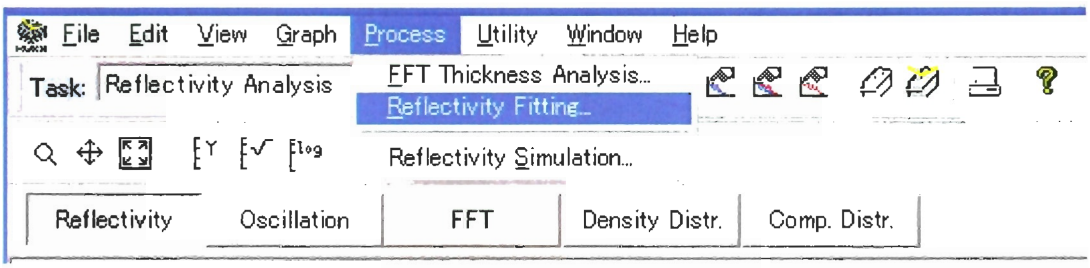
Open "Au.gxr" file saved in following path, C:¥Program Files¥Rigaku¥GXRR3¥Data.

Designed model ; Au (20nm) / Si substrate



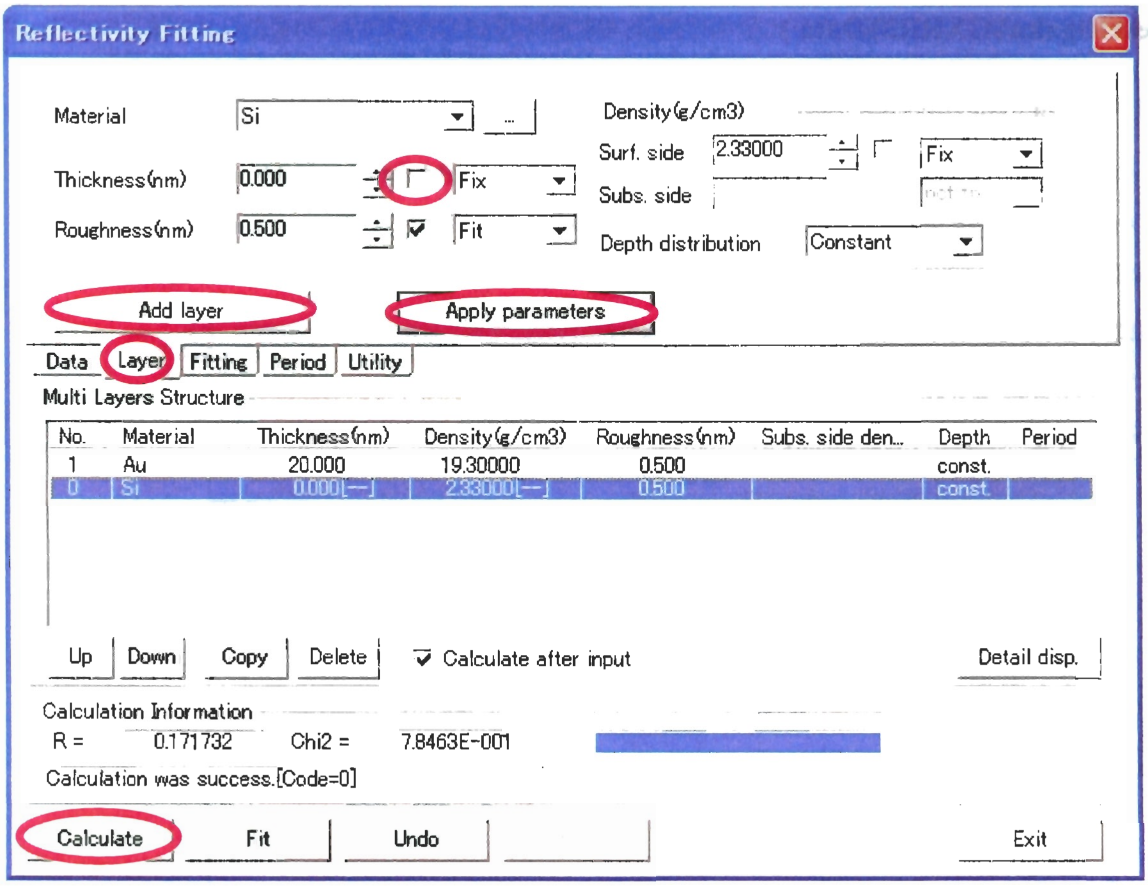
3-4-2 Set the designed model

First, Select [Process] – [Reflectivity fitting] of click (icon) to make a structure model of layer.

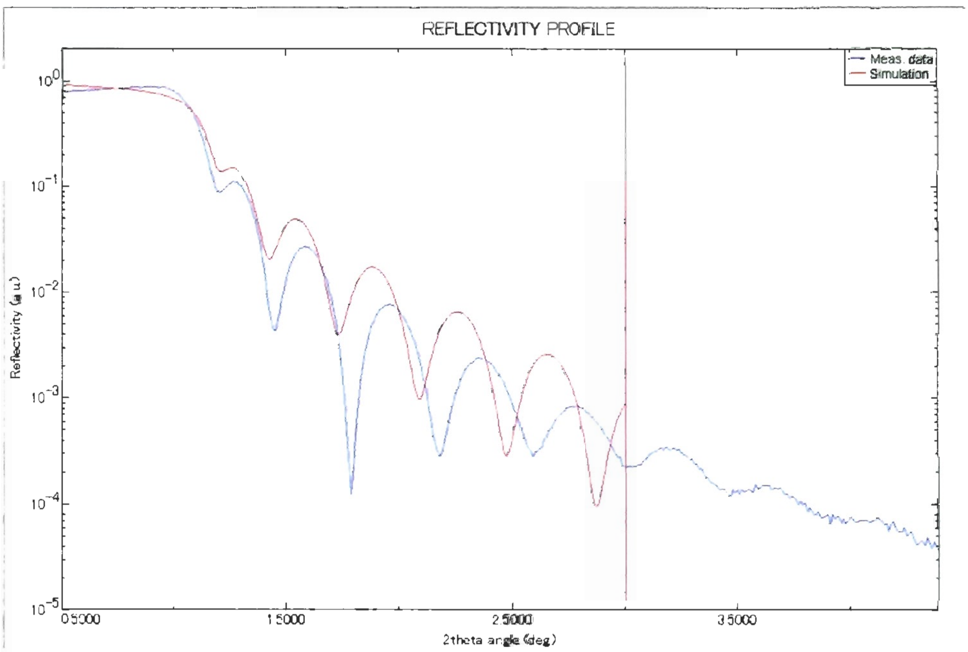


Select [Material] in [layer] tab, then select **Si** and click [Add layer]. Next, select **Au** and input [Density], [Thickness] and [roughness] as the designed values.

Remove [Thickness], [Roughness], [Density] checkbox of **Au layer**, [Roughness] checkbox of **Si substrate** and [Apply parameters] to set as fitting parameters.



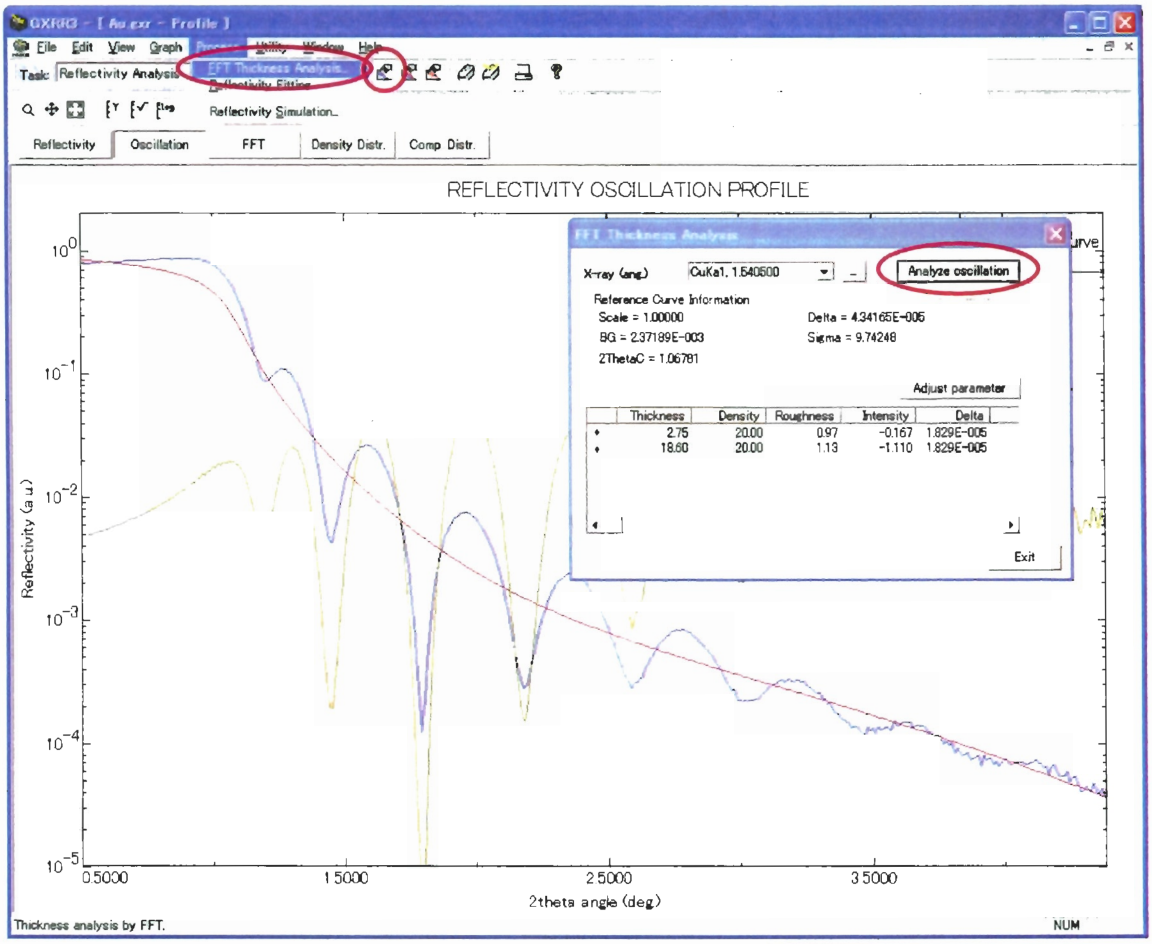
Click [Calculate] to display the calculated profile. The difference between calculated and measured profile indicates that those parameters used in the calculation differ from actual values.



3-4-3 Estimation of film thickness by Fourier transform

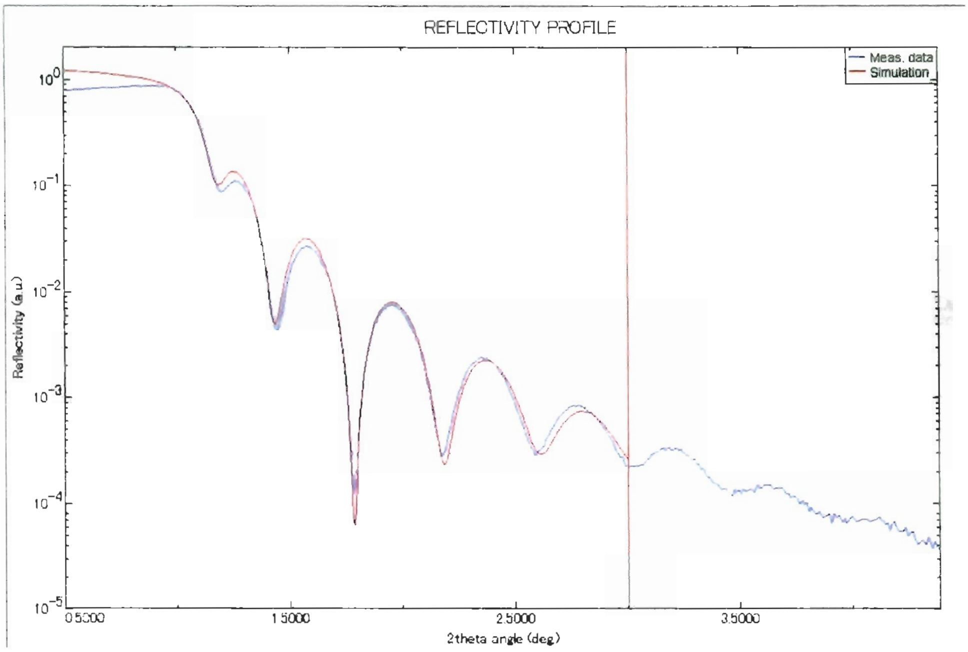
Film thickness is estimated by Fourier transform of oscillation components appearing on an XRR profile. The result is reliable in the case of single-layer film, and it is also useful for multi- layer samples because it still gives some hints for XRR analysis.

[FFT Thickness] dialogue appears by choosing [Process] – [FFT Thickness Analysis] or click . Click [Analyze oscillation]. The oscillation components are separated as follows.



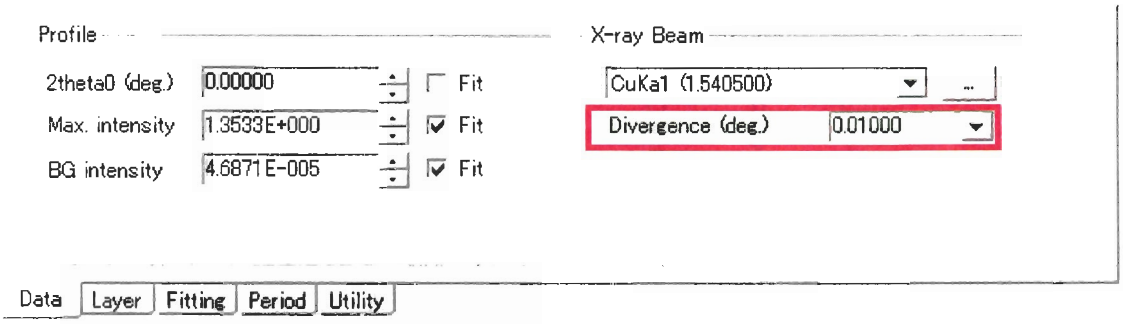
This oscillation is transformed to film thickness by the period of the oscillation and the electron density of the film calculated from chemical formula and the density. The results of FFT analysis (Thickness, Density, Roughness) show on FFT Thickness Analysis dialog.

Click [Adjust parameter] to adjust the results of FFT analysis to the layer model.

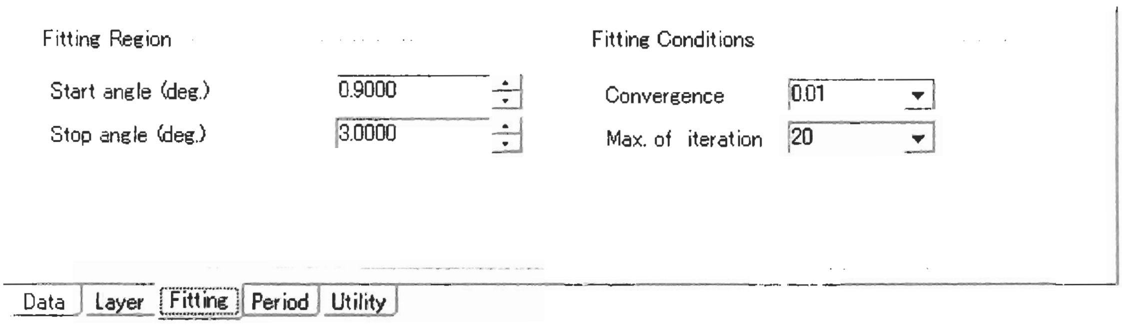


3-4-4 Profile Fitting

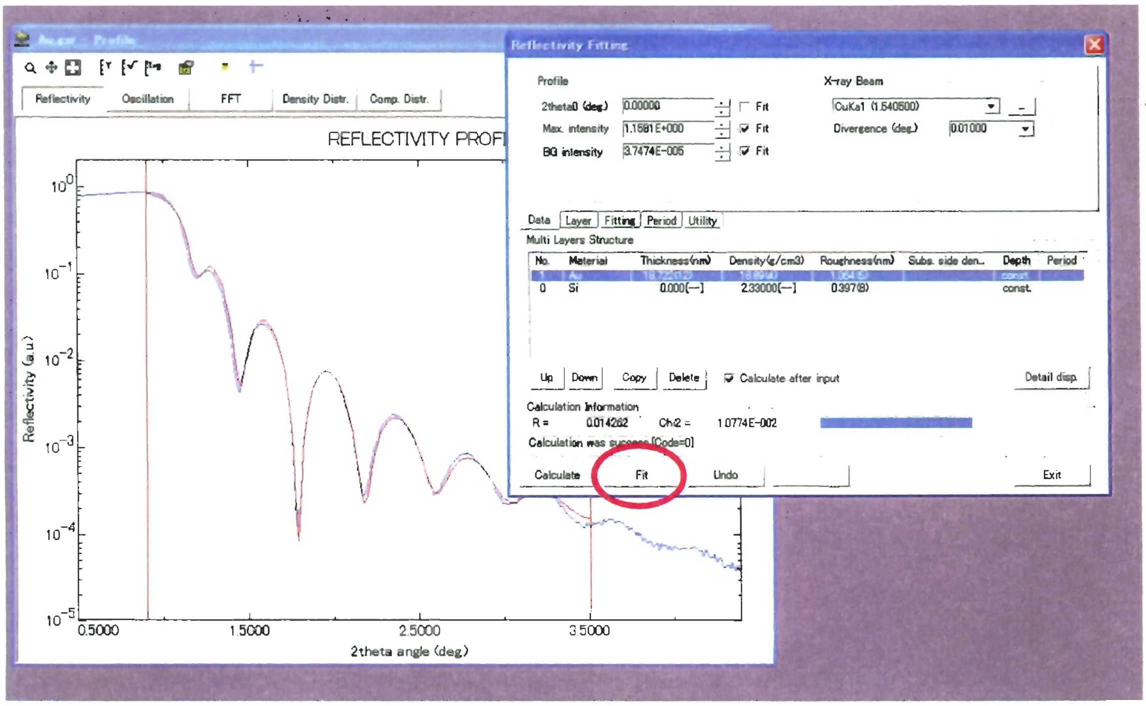
Input [Divergence] corresponding to optical system in [Data] tub.



Set [Fitting Region] to decide the fitting range. Ordinary, input [Start] to the critical angle and [Stop] to the angle that intensity becomes background level.



Click [Fit] to start profile fitting. Parameters which have checkmark in their checkbox are refined. Profile fitting will be considered to finish when necessary parameters converge and R-value decreases to ~0.04.


Reflectivity Fitting
Reflectivity
Oscillation
FFT
Density Distr.
Comp. Distr.
REFLECTIVITY PROFI
Profile
2theta0 (deg.)
0.00000
Fit
Max. intensity
1.1581E+000
BG intensity
3.7474E-005
X-ray Beam
CuKa1 (1.540600)
Divergence (deg.)
0.01000
Data
Layer
Fitting
Period
Utility
Multi Layers Structure
No.
Material
Thickness(nm)
Density(g/cm3)
Roughness(nm)
Subs. side den..
Depth
Period
0
Si
0.000[---]
2.33000[---]
0.397(8)
const.
Up
Down
Copy
Delete
Calculate after input
Detail disp.
Calculation Information
R =
0.014262
Chi2 =
1.0774E-002
Calculate
Fit
Undo
Exit
Reflectivity (a.u.)
2theta angle (deg)
0.5000
1.5000
2.5000
3.5000