

Appendix to the manual of the ROD program (experimental version): Robach's extension

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0) Introduction

0.1) PRESENTATION AND ORGANIZATION

This manual describes the ROBACH extension of the ROD program. This is only a partial manual: for the standard features of the ROD program, one should refer to Elias Vlieg's "a concise ROD manual", which gives the basics of the program.

A section at the end of the manual reviews some of the bugs that occur when running the program (those bugs belong to the standard version and are not crucial for the calculation).

In this manual, we first explain "theoretically" the features of the extension and the kind of physical problems those features enable to treat. We start by the simple features ("single" parameters), and then we continue with the more complicated features (a model that enables to automatically generate several atomic planes).

A summary of the ROD menus corresponding to this extension is given at the end of this manual ("In practice" section). For those who want to try without reading the manual: the extension can be accessed from the main menu by the command "EXT ROB".

In the section concerning substrate + adsorbate systems (section 2), there are also some useful formulas about roughness.

0.2) Contributors

Contributors to ROD are so numerous that it is difficult to name them all: this extension results from the work of persons of the CEA-Grenoble (G. Renaud, P. Guénard), the staff of ID3, and myself.

0.3) Disclaimer

Don't expect the manual to be perfect. We try to keep it up-to-date with the versions. Please send your comments to robach@esrf.fr if you detect any errors or have ideas to make the version more complete.

0.4) Possible improvements

For a long time we have treated the interdiffusion between adsorbate and substrate indirectly, through a varying roughness of the substrate. A varying roughness of the substrate (as deduced from the CTRs) could arise from many different phenomena:

- either from a real exchange of adsorbate and substrate atoms, with or without a change in the lateral lattice parameter of the substrate planes concerned,
- or just from the inhomogeneous strain field induced in the substrate by strained adsorbate islands, without any atomic exchanges.

It would be interesting to examine those different cases, and see if we can use the measurements of the CTRs to distinguish between them: they could for example produce an effect on the CTRs that has a different L or $q_{//}$ variation depending on the phenomenon that is involved.

Other ideas:

- introduce concentration = $f(z)$ profiles for the segregation of an impurity to the surface of a crystal.

- introduce the Distorted Wave Born Approximation to treat correctly the refraction problems. Of course, in order to use this kind of refinement in the analysis of the CTRs, one would need to know in advance the profile of electronic density in the deposit.
- add a menu with growth models to fit the growth oscillations in the specular or the CTRs.

0.5) A few remarks concerning the “extended” model

i) something you could ask: “why do you insist on fitting the truncations rods of the substrate in presence of an adsorbate, while you could get most of the morphological information by measuring the reflectivity, and with the reflectivity you would not have to care if your adsorbate is strained or relaxed in the surface plane?”

The answer is the following: of course, if you can measure the “extended” reflectivity (i.e. the (00L) rod up to high L), it is certainly better to use it to get the thickness and the roughness of the deposit (or more generally the profile of electronic density along z in the adsorbate). In fact the model we describe can also be used to fit reflectivity profiles, although programs containing refinements such as the Distorted Wave Born Approximation are certainly better.

Fitting the substrate CTRs other than the (00L) is interesting:

- when you can’t measure the extended reflectivity. This is the case for the light oxide systems (MgO, Al₂O₃) and also for clean Si (001) surfaces: the noise from the bulk becomes huge as soon as the incident angle is larger than the critical angle, so that the high L part of the reflectivity is not measurable.
- when you want information such as:
 - the adsorption site (its lateral coordinates, the z-coordinate you can get from the reflectivity)
 - the degree of in-plane relaxation of your deposit (for example: is there a pseudomorphic part?). Note that for detecting a very small amount (0.2 ML) of relaxed adsorbate growing as islands, the measurement of the substrate CTRs is much more sensitive (coherent adding + narrow peaks) than the measurement on the peaks of the relaxed adsorbate (incoherent adding + very broad peaks).

ii) the idea that *relaxed* adsorbate can produce modifications of the substrate CTRs (section 2.4) is something rather new. It was already used by Toney et al for Tl on Ag(111) in a slightly different context (“Surface x-ray scattering measurements of the substrate-induced spatial modulation of an incommensurate adsorbed monolayer”, M.F. Toney et al, PRB 42(9) (1990) 5594), but I don’t know of any other paper using this idea for surface diffraction.

This idea is potentially interesting because it shows that the presence of interference on the substrate CTRs does not necessarily imply that there is any pseudomorphic adsorbate.

0.6) New features (versions after April 1st

NEW 2001

As a general rule the new features are highlighted in yellow (at least the title), to make them easier to locate. In this section we give only a summary of the new features. The details are dispersed into the several highlighted sections of the manual.

Additions for versions after April 1st 2001:

- Several remarks about the problem of fitting the crystal truncation rods of Si(001), Sb/Si(001) and Ge/Sb/Si(001): some progress in the question of how to treat the roughness for Si(001) and more generally for surfaces with several non-equivalent terminations. This leads to the addition of a new column NLAYER (optional) in the data file, and to the use of SIGMA (rms roughness) instead of BETA in the roughness calculation. (08/11/00)
- A second occupancy serial number for each atom, which enables to treat interdiffusion between two planes. (08/11/00)
- Some more comments in the parts about the effect of relaxed vs. pseudomorphic adsorbate on the substrate truncation rods (03/01/01).
- A new flag in the menu EXT ROB SET CALC that enables to multiply or not the surface contribution by the roughness factor of the bulk (16/02/01).

1) The simple features

1.1) The LSHIFT

This is a fitting parameter that enables you to compensate for slight misalignments of the CTRS around a Bragg peak (for example you have a CTR with a Bragg peak at $L=1$ and $I(L=1.05) \gg I(L=0.95)$ while physically you expect the intensity to be symmetric around the Bragg peak)

The way this parameter works is that ROD tries to fit the data points:

$F_{dat}(H_{dat}, K_{dat}, L_{dat})$

using the theoretical structure factor:

$F_{th}(H_{dat}, K_{dat}, L_{dat} + LSHIFT)$

instead of:

$F_{th}(H_{dat}, K_{dat}, L_{dat})$.

Of course if your data are always well-aligned you don't need the LSHIFT, but this is not always the case for me.

For the moment there seems to be a bug when using the LSHIFT: the present version of ROD does not give the same results as older versions. So one has to take this parameter with caution. Hopefully the bug will be corrected soon.

1.2) The SCALE3 (SC3)

This is also a fitting parameter, which enables you to apply to the "fractional order" points of a reconstruction a different scale factor than for the CTRS.

With this option you can do a simultaneous fit of the CTRS and of the in-plane reconstruction points, even if they have been measured in different conditions.

The theoretical structure factor calculated by ROD will be:

$SCALE * (roughness\ factor) * F_{th}$ for the CTRS

and

$SCALE * SCALE3 * F_{th}$ for the fractional orders.

This renders the fractional orders insensitive to the value of the roughness.

Note that the SCALE3 parameter differs from the SCALE2 parameter. When using SCALE2, for the fractional orders, the theoretical structure factor is:

$$SCALE*SCALE2*roughness_frac*Fth$$

where *roughness_frac* is a roughness factor specific to fractional orders. See calc_roughness routine in calc.c for the details. For the “approximated beta” roughness model:

$$roughness_frac = [(1-\beta)/(1+\beta)]^{1/2}$$

When using the ROBACH extension, the SCALE2 parameter is not used in the calculation.

In practice, in your data file, you have to put positive LBRAGG values for the CTR points and negative LBRAGG values for the fractional orders points. This is what enables ROD to recognize which kind of point it is dealing with.

1.3) The SURF2FRAC

This parameter, which is the fraction of the reconstructed surface occupied by the second surface unit cell, already exists in the standard version as a fitting parameter. The only new thing is that now it has the “range checking” option.

1.4) Additional remarks

The 3 above parameters have the "range checking" option in the fits, and they can be modified either in the EXT ROB SET PAR menu or in the standard FIT menu.

1.5) The ERF roughness model

The standard version of ROD includes only one approximate roughness model (the β^n model of I.K. Robinson). This model was called “approximate beta model” in the program and you could select it in the menu SET CALC ROUGH. Several “exact” roughness models are also available in this menu.

Here we added a second approximate roughness model. This model (programmed by P. Guénard) corresponds to a Gaussian distribution of the terrace heights, i.e. to an erfc(z) occupancy profile of the different planes. This second roughness model can be selected in the EXT ROB SET PAR menu by the command “erf yes”.

For the ERF model, the formula that ROD uses for the roughness factor is:

$$roughness = rough\left(\frac{l - l_{BRAGG}}{nlayers}\right)$$

where

$$rough(x) = \frac{1 + \sum_{n=1}^{Nmax} 2 \cdot e^{-\frac{1}{2}\left(\frac{n}{\beta}\right)^2} \cdot \cos(2\pi \cdot n \cdot x)}{\sum_{n=0}^{Nmax} 2 \cdot e^{-\frac{1}{2}\left(\frac{n}{\beta}\right)^2}} \quad (\text{needs to be checked})$$

where nlayers is the number of non-equivalent layers in the bulk unit cell (the one entered in SET CALC), l is the current perpendicular Miller index, l_{BRAGG} is the value

of 1 for one of the Bragg peaks on the truncation rod, and Nmax is the integer part of $10 \times \beta$.

RMS roughness:

For the β^n model, the formula giving the rms roughness is:

$$\sigma_{RMS} = \frac{\sqrt{\beta}}{1-\beta} \cdot \frac{c_s}{nlayers}$$

where $0 < \beta < 1$ and c_s is the lattice parameter of the substrate along z (the one given in the .bul file).

For the ERF model, the rms roughness is given by:

$$\sigma_{RMS} = \frac{c_s \cdot \beta}{Nlayers} \quad (\text{needs to be checked})$$

1.6) The ZCONST, Z2CONST and NZ2DIS

These features are identical to the XCONST, X2CONST and NX2DIS (YCONST, Y2CONST and NY2DIS) that exist for the X and Y in-plane displacement parameters. They enable to use the same displacement (the same serial number) for different atoms with different multiplication factors. This allows for example to link the z-displacement of an atom to its x or y displacement, or to link the z-displacements of the different atoms to make them vary sinusoidally as a function of the starting x-position etc...

To describe the way the parameters work, it is easier just to give the corresponding program lines (which belong to the source file set.c):

```

/*****/
void update_model(void)
/*****/
/* Update the parameters of the model */
{
  int i;

  for (i = 0; i < NSURF + NSURF2; i++)
  /* for each atom i belonging to the surface unit cell */
  {
    /* starting value of x for this atom */
    XSFIT[i] = XS[i];
    /* add the first x displacement */
    if (NXDIS[i]>0) XSFIT[i] += XCONST[i] * DISPL[NXDIS[i]-1];
    /* add the second x displacement */
    if (NX2DIS[i]>0) XSFIT[i] += X2CONST[i] * DISPL[NX2DIS[i]-1];

    YSFIT[i] = YS[i];
    if (NYDIS[i]>0) YSFIT[i] += YCONST[i] * DISPL[NYDIS[i]-1];
    if (NY2DIS[i]>0) YSFIT[i] += Y2CONST[i] * DISPL[NY2DIS[i]-1];

    ZSFIT[i] = ZS[i];

    if (robach_flag!=0)
    {
      if (NZDIS[i]>0) ZSFIT[i] += ZCONST[i] * DISPL[NZDIS[i]-1];
      if (NZ2DIS[i]>0) ZSFIT[i] += Z2CONST[i] * DISPL[NZ2DIS[i]-1];
    }
  }
}

```

```

else
    if (NZDIS[i]>0) ZSFIT[i] += DISPL[NZDIS[i]-1];

```

As can be seen in the program, this option works only when using the Robach's extension (robach_flag=1).

1.7) The NOCCUP2 parameter: treating interdiffusion

Now in the .fit file, each atom "i" can have two different serial numbers NOCCUP[i] and NOCCUP2[i] for the occupancy (the second one, NOCCUP2, is optional). The final value of the occupancy for atom "i" will be the difference between the two occupancies. In the update_model routine of set.c, we have now:

```

if (robach_flag)
{
    OCCUPTOT[i] = 1.0;
    if (NOCCUP[i] > 0)
        OCCUPTOT[i] = OCCUP[NOCCUP[i]-1];
    if (NOCCUP2[i] > 0)
        OCCUPTOT[i] += -OCCUP[NOCCUP2[i]-1];
}

```

Normally, for a given atom i, one can put (Dec 11th 2001) the first serial number (NOCCUP[i]) at zero and the second serial number (NOCCUP2[i]) at a non-zero value, so as to have:

```
OCCUPTOT[i] = 1.0 - OCCUP[NOCCUP2[i]-1]
```

This works also when using the "PG-extended model" (described later).

This is interesting for treating interdiffusion. Let us consider for example the case of one monolayer of Ge grown on Si. We want to allow for an interdiffusion between the Ge layer (supposed to be flat) and the last Si plane.

In the case without interdiffusion the model would be:

atom 1:

Si (xs, ys, zs) = (0, 0, 0.5)

atom 2:

Ge (xs, ys, zs) = (0, 0.5, 1.0) NOCCUP[2]=1

where NOCCUP[2]=1 allows for a partial filling (=occ1) of the Ge plane.

In the case with interdiffusion, the model would be:

atom 1:

Si (xs, ys, zs) = (0, 0, 0.5) NOCCUP[1]=1 NOCCUP2[1]=2

atom 2:

Ge (xs, ys, zs) = (0, 0.5, 1.0) NOCCUP[2]=3 NOCCUP2[2]=2

atom 3:

Ge (xs, ys, zs) = (0, 0, 0.5) NOCCUP[3]=2

atom 4:

Si (xs, ys, zs) = (0, 0.5, 1.0) NOCCUP[4]=2

By fixing OCCUP[1]=occ1=1.0, we have:

- in the first plane: occ(Si)=1-occ2, occ(Ge)=occ2

- in the second plane: $occ(Si)=occ2$, $occ(Ge)=occ3-occ2$.

This allows both an interdiffusion between the Ge and the Si ($occ2$ is the amount of Si exchanged between the two planes) and a variable total amount of Ge ($occ3$).

2) More complicated now: dealing with several layers of adsorbate

2.1) Introduction

All that follows is only interesting in the following case:

- you are trying to fit crystal truncation rods of a substrate
- there is an adsorbate on top of your substrate
- this adsorbate occupies more than one atomic plane.

Let's take an example:

You have deposited on Si(001) an amount of Ge equivalent to five (001) monolayers. You see oscillations on the CTRs of the Si, and you are looking for a model to fit that.

The first idea is to say that the oscillations are created by PSEUDOMORPHIC Ge, i.e. Ge that has the SAME in-plane lattice parameter as the Si (this Ge is strained by the Si). This is not necessarily the right interpretation (see section 2.4) but this is what we will assume from here to section 2.3 included.

With this hypothesis of pseudomorphic Ge, you are facing several questions:

- which fraction of the deposited Ge is effectively pseudomorphic?
(parameter occ -pseudo)
- how many planes is this pseudomorphic Ge distributed over?
(parameter thickness-pseudo)
- what is the interlayer distance between the planes of pseudomorphic Ge, and does this distance vary with z (the height of the plane with respect to the interface)?
(parameters dz -pseudo and dz -relax-pseudo)
- what is the distance between the last layer of Si and the first layer of pseudomorphic Ge?
(parameter d -interfacial)
- do all the planes of the pseudomorphic Ge have the same occupancy or is there a certain occupancy profile in z ?
(parameter roughness-pseudo)
- where are the Ge atoms in the first pseudomorphic plane sitting with respect to the Si lattice?
(parameter adsorption-site)

As you can see, even with a very simple description of the deposit, there are already many parameters.

If you imagine that you have to build your surface model Ge atom by Ge atom, with a z -displacement and an occupancy for each atom, and this over 10 Ge planes, you immediately realize that your fit will never get anywhere.

2.2) The "PG - extended model": PSEUDOMORPHIC adsorbate

2.2.1) Short description

The "PG - extended model" enables to solve the problem we just described. This model has been created and put into ROD by Pascal Guénard (a former Ph.D. student of G. Renaud at the CEA, now at IBM).

With this model, instead of having to deal with a dozen of z-displacements and occupancies in the fits, you deal directly with the parameters:

- occ-pseudo
- thickness-pseudo
- dz-pseudo
- dz-relax-pseudo (optional)
- d-interfacial
- roughness-pseudo

The parameter "adsorption site" is usually adjusted by hand, although in theory it should be possible to make it vary in the fits using x and y displacements.

2.2.2) How to build the model in ROD: the EXT ROB SET ADSORBATE menu

We describe below the way you build the "extended" model in ROD, using the example of Ag (FCC) deposited on MgO (001) (FCC, NaCl structure). This is of course only a classroom example since in reality Ag is never pseudomorphic on MgO.

Examples showing the exact syntax of the ROD commands are given in section 3.

2.2.2.1) The .BUL file

The .bul file is the same as in the standard version, except that one can add a fifth column indicating the serial number of the occupancy parameter for the bulk atoms.

For Ag/MgO: we use the bulk unit cell of MgO, and the oxygen atoms of the last MgO plane are at the reduced coordinates: $(x\ y\ z) = (0\ 0\ 0)$ and $(0.5\ 0.5\ 0)$

2.2.2.2) The .FIT file

In the .fit file, we put only the first plane of Ag, that is two Ag atoms, that we put above the oxygen atoms of the last MgO planes, at $(x\ y\ z) = (0\ 0\ 0.5)$ and $(0.5\ 0.5\ 0.5)$.

For creating the parameter d-interfacial, we give to these two Ag atoms the same z-displacement, with the serial number 1 (dz1). After creating the other Ag planes, dz1 will represent a "block" z-displacement of all the Ag atoms, not only a displacement of the 2 Ag atoms in the first plane.

For creating the parameter occ-pseudo, we give to the two Ag atoms in the .fit file the same occupancy parameter, with the serial number 1 (occ1). After creating the other Ag planes, depending on which model you select for representing the occupancy profile, occ1 will represent either the total amount of pseudomorphic Ag (expressed in monolayers) or the occupancy of the first Ag plane.

2.2.2.3) The .PAR or .MAC file

Now we start building the other Ag planes.

The idea is to take EACH Ag atom in the .fit file and to “extend” it over several planes, i.e. you create automatically new atoms that are (more or less) above the initial atom. This looks very simple but it takes quite a number of parameters to define where you want to put the new atoms.

Let’s start by extending the Ag atom that is number 1 in the .fit file. Note that the atoms in the .fit file are numbered from 1 to N (and not from 0 to N-1).

In ROD you go to the menu “EXT ROB SET ADS”, then to the sub-menu “EXTENDED”. This asks you first which atom you want to “extend”, so you enter “1”, and you arrive in the menu EXT.ROB.SET.ADS.EXT>. In this menu, you have a series of parameters to set, which are described below.

2.2.2.3.1) MODE parameter (yes/no)

This parameter serves to tell ROD to “extend” or not the atom number n (here number 1). Of course we answer “yes”.

2.2.2.3.2) NPLANS parameter

In extending the Ag atom number 1, you have first to define an upper limit for the number of Ag planes that will be present in your model. All those planes will not necessarily be occupied by Ag atoms, they just have to be there to allow the Ag thickness to vary without having to modify the model by hand. Typically we take NPLANS=20.

2.2.2.3.3) NLAYERS parameter

This parameter plays for the adsorbate a role similar to the nlayers used in the calculation of the substrate roughness factor (SET CALC menu). It tells ROD how many non-equivalent planes there are in the adsorbate unit cell along z. For Ag (001), there are 2 non-equivalent planes so we take NLAYERS=2. For Ge (001), NLAYERS=4.

If the adsorbate has a structure different from that of the substrate, the NLAYERS defined in the EXT ROB SET ADS EXT menu for the adsorbate is not necessarily the same as the nlayers defined in SET CALC for the substrate.

2.2.2.3.4) XSHIFT, YSHIFT parameters

Now that ROD knows how many non-equivalent layers there are in the adsorbate unit cell, you have to tell it, layer by layer, where the adsorbate atoms are in those non equivalent layers.

For Ag (001), we have NLAYERS=2. In layer number 1, the Ag atom is at the position given in the .fit file. In layer number 2, the Ag atom will be shifted by (XSHIFT, YSHIFT)=(0.5, 0) with respect to the atom in layer 1.

2.2.2.3.5) The ZDIS and ODIS parameters

Now that ROD has the x y positions of the Ag atoms for the different layers and the maximum number of layers, you have to tell it where to put the planes in z, and which occupancy to give them. The idea is of course to avoid entering all those z-positions and occupancies by hand. For this, you have the choice between a number of possible occ(z) occupancy profiles, and between a number of possible $\Delta z(z)$ interlayer distance profiles. Each of the profiles is parameterized with one to four parameters. Those parameters are the ones you do the fit on.

The definition of the occ(z) and $\Delta z(z)$ profiles is done separately in the EXT ROB SET ADS FUNC menu that will be described in details below (section 2.2.4). Each occ(z) and $\Delta z(z)$ function that you enter in ROD will have a separate serial number. The attribution of this serial number is done in the EXT ROB SET ADS FUNC menu.

In the EXT ROB SET ADS EXT menu, all you have to do is to tell ROD the serial numbers of the occ(z) and $\Delta z(z)$ functions that you want to use for this particular extended atom. This is done via the commands ODIS and ZDIS.

2.2.2.3.6) Other parameters

The other parameters that you find in the EXT ROB SET ADS EXT menu, such as NSTART, ORIGIN and ZSHIFT, are more “specialized” parameters, which are designed for specific cases. Their use will be described in details in section 2.2.4) which presents the different possible occ(z) and $\Delta z(z)$ profiles.

Reasonably safe choices are :

NSTART=0

ORIGIN = the number (in the .fit file) of the atom you are extending

ZSHIFT = 0. (this works for Ag/MgO but not for Ge/Si(001)).

2.2.3) The occ(z) and $\Delta z(z)$ profiles

2.2.3.1) The EXT ROB SET ADS FUNC menu

In the EXT ROB SET ADS FUNC menu, you can define several occupancy functions and interlayer distance functions, each characterized by a serial number (1 to N1 for the occ(z) functions, 1 to N2 for the $\Delta z(z)$ functions), a type of profile, and one to four parameters. The different types of profiles and their parameters are described below.

For creating an occ(z) function, you enter “oadd”, then you enter its serial number.

For creating a $\Delta z(z)$ function, you enter “zadd”, then you enter its serial number.

For changing the parameters of an occ(z) function (resp. of a $\Delta z(z)$ function) you enter “opar” (resp. “zpar”).

2.2.3.2) Simplifying assumptions

In order to simplify the description that follows, we will assume that for the extended atom number i:

NSTART = 0

ORIGIN = i
 ZSHIFT = 0.

In section 2.2.3.7), we will describe the influence of the parameters NSTART and ORIGIN on the $\Delta z(z)$ function.

The ZSHIFT parameter serves to change the origin of the occ function: an atom located at height z will have the occupancy $\text{occ}(z - \text{ZSHIFT})$ instead of $\text{occ}(z)$. The interest of the ZSHIFT parameter is described in section 2.3.4) for the case of Ge/Si (001).

2.2.3.3) INTERLAYER DISTANCE profiles

In this part we describe the various $\Delta z(z)$ profiles that are available.

The different atomic planes (one atom per plane) created by extending atom number i are numbered with an integer n (positive or negative) that varies either from 0 to (NPLANS-1) or from (-NPLANS+1) to (NPLANS-1) (depending on which $\text{occ}(z)$ function is used). We name:

- $z(n)$ the z -position of the n^{th} plane,
- $\Delta z(n)$ the interlayer distance between plane $(n-1)$ and plane n .

For the $\Delta z(n)$ plots we will consider only the cases where n goes from 0 to (NPLANS-1).

For all the profiles, with the option (ORIGIN=i), we have:

$$z(0) = \text{ZSFIT}[i] \tag{Z.1}$$

where ZSFIT(i) is the z -position of atom i . (the atom $n=0$ is simply atom i).

The interlayer distance and the z -positions of the different planes are calculated in a recurrent way, with equations such as:

$$\Delta z(n) = z(n) - z(n-1) = f(z(n-1)) \tag{Z.2}$$

and

$$z(n) = z(n-1) + \Delta z(n) \tag{Z.3}$$

2.2.3.3.1) NO relaxation

This profile has one parameter: dz_{∞} .

The interlayer distance is uniform:

$$\Delta z(n) = dz_{\infty} \tag{Z.4}$$

and the z -position is simply given by:

$$z(n) = z(0) + n \cdot dz_{\infty} \tag{Z.5}$$

2.2.3.3.2) EXP relaxation

This profile has three parameters:

- dz_{∞} , which is the value that Δz tends toward for large n 's
- dz_0 , which is the value of Δz for $n=1$
- sigmaz , which is the “attenuation constant” of the Δz function.

This profile normally works with:

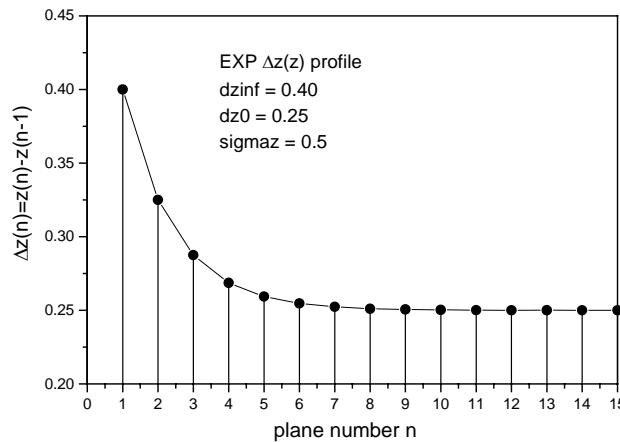
$$0 < \text{sigmaz} < 1 \quad (\text{Z.6})$$

The interlayer distance is given by:

$$\Delta z(n) = dz_{\infty} + (dz_0 - dz_{\infty}) \cdot (\text{sigmaz})^{n-1} \quad (\text{Z.7})$$

and the z-position is given by :

$$z(n) = z(0) + n \cdot dz_{\infty} + (dz_0 - dz_{\infty}) \cdot \frac{\text{sigmaz}^n - 1}{\text{sigmaz} - 1} \quad (\text{Z.8})$$



2.2.3.3.3) ERF relaxation

This profile has four parameters:

- dz_{∞} , which is the value that Δz tends toward for large n 's
- dz_0 , which is the value of Δz for $n=1$
- sigmaz , which is the “extension” of the Δz function
- z_{meanz} , which is the “central” z value of the erf function.

There are two versions for the erf relaxation:

1) in the first version, the parameters sigmaz and z_{meanz} of the $\Delta z(z)$ function are the same as the parameters sigmao and z_{meano} of the $\text{occ}(z)$ function having the same serial number as the $\Delta z(z)$ function. The fit is done on sigmao and z_{meano} , and of

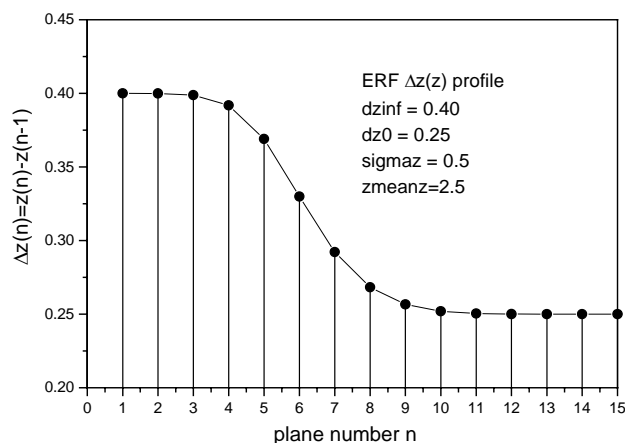
course an $occ(z)$ function with the same serial number as the $\Delta z(z)$ function has to be created.

2) in the second version, $sigmaz$ and $zmeanz$ are independent parameters that are not linked to the parameters of the $occ(z)$ functions.

The interlayer distance is given by:

$$\Delta z(n) = dz_{\infty} + (dz_0 - dz_{\infty}) \cdot \frac{1}{2} \cdot \operatorname{erfc} \left(\frac{z(n-1) - zmeanz}{\sqrt{2} \cdot sigmaz} \right) \quad (Z.9)$$

The z -position has no simple form and is deduced in a recurrent way using equation (Z.3).



2.2.3.4) OCCUPANCY profiles

2.2.3.4.1) Parameters common to all the profiles

Now that ROD knows the z -positions of the different atoms created by extending atom number i , it can calculate their occupancy from the $occ(z)$ function that we have attributed to this extended atom.

All the occupancy profiles have among their parameters one occupancy parameter, which is the occupancy $occ(snocc(i))$ that you gave in the .fit file (through its serial number $snocc(i)$) to the atom number i that you are extending. In our example on Ag/MgO, this occ parameter has the serial number 1 so we call it $occ1$.

For all the occupancy functions except the last one (section 2.2.4.3.e), $occ(snocc(i))$ represent the TOTAL amount (in monolayers) of atom number i , including all the atoms created by extending atom i .

The formulas that ROD uses are:

- for the erf and exp occupancies:

$$occ(snocc(i)) = \sum_{n=0}^{n=NPLANS-1} occ(n,i) \quad (O.1a)$$

- for the gauss occupancy:

$$occ(snocc(i)) = \sum_{n=-NPLANS+1}^{n=NPLANS-1} occ(n,i) \quad (O.1b)$$

where $occ(n,i)$ is the occupancy of the n^{th} -atom created by extending atom number i . This n^{th} atom sits in the layer number n , that has a certain z -position $z(n,i)$, and:

$$occ(n,i) = occ(z(n,i)) \quad (O.2)$$

For the last occupancy function (erf occupancy with fit on the first plane occupancy), $occ(snocc(i))$ represent directly the occupancy of atom i , i.e. the occupancy of the first plane of extended atoms of type i .

ROD uses the formula:

$$occ(snocc(i)) = occ(0,i) \quad (O.3)$$

We will now describe the different occupancy functions.

2.2.3.4.2) UNIFORM occupancy:

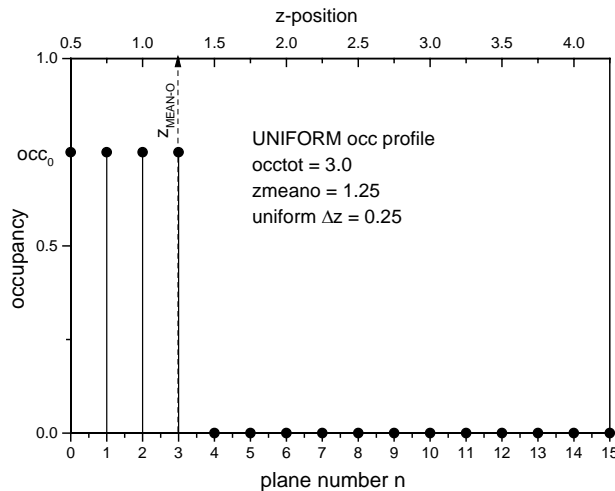
Here all the layers have the same occupancy under a certain z , and then a zero occupancy above. This profile has two parameters:

- $occtot$, which was already mentioned ($=occ(snocc(i))$)
- z_{MEANO} , which acts as the thickness of the adsorbate.

ROD uses the formula :

$$\begin{aligned} \text{if } (z \leq z_{MEANO}) \quad & occ(z) = occ_0 \\ \text{else} \quad & occ(z) = 0 \end{aligned} \quad (O.4)$$

where occ_0 is derived from the condition on the sum of the occupancies expressed in equation (O.1a).



2.2.3.4.3) ERF occupancy with fit on total occupancy

This profile has three parameters:

- $occtot$, which was already mentioned ($=occ(snocc(i))$)
- z_{MEANO} , which acts as the average thickness of the adsorbate
- σ_{MAO} , which acts as a roughness of the surface of the adsorbate.

Note that the complex structure factor of the adsorbate is always multiplied by the roughness factor of the substrate. Therefore, the roughness σ_{MAO} acts as an additional roughness of the adsorbate, i.e. a roughness that the adsorbate has in addition to the “automatic” roughness it gets from following the steps profile of the substrate.

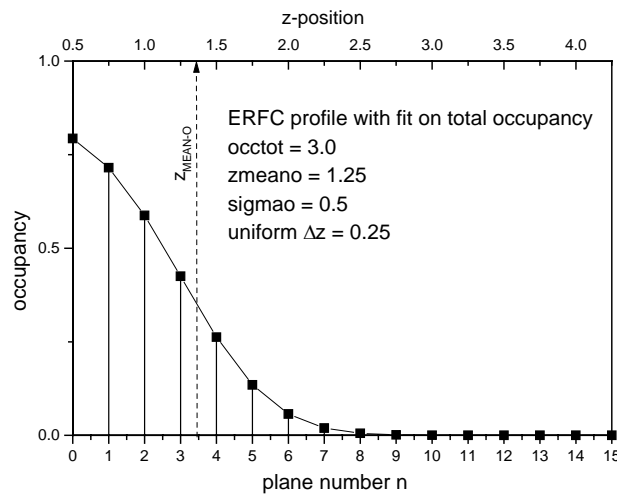
$\sigma_{MAO} = 0$ corresponds to a growth that is either layer by layer or in islands with vertical edges. Note that in this case, the erf profile is the same as the “uniform occupancy” profile.

$\sigma_{MAO} \neq 0$ corresponds to the « 3D » and Stranski-Krastanov growths, involving islands with non-vertical edges and possibly complete layers.

The formula used by ROD in this case is:

$$occ(z) = occ_0 \cdot \frac{1}{2} \cdot erfc\left(\frac{z - z_{MEANO}}{\sqrt{2} \cdot \sigma_{MAO}}\right) \quad (O.5)$$

where $erfc$ is the complementary error function, and occ_0 is derived from the condition expressed in equation (O.1a).



2.2.3.4.4) GAUSS occupancy

This kind of occupancy profile can be useful for treating the case where there is intermixing between the substrate and the adsorbate at the interface.

Like the preceding one, this profile has three parameters:

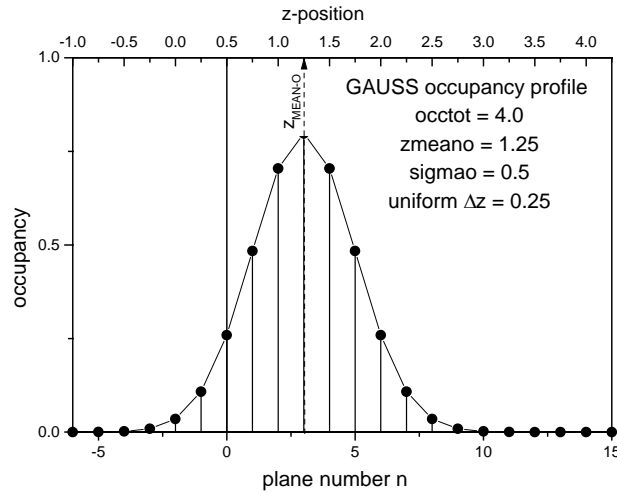
- $occtot$
- z_{MEANO}
- σ_{MAO} ,

that play exactly the same role as before, except for the exact form of the function.

Note that when using this function, the adsorbate atoms are distributed over $(2 \cdot NPLANS - 1)$ planes instead of $NPLANS$ planes, and that n can take negative values. The formula used by ROD in this case is:

$$occ(z) = occ_0 \cdot \exp \left[\left(\frac{z - z_{MEANO}}{\sqrt{2} \cdot \sigma_{MAO}} \right)^2 \right] \quad (O.6)$$

where occ_0 is derived from the condition expressed in equation (O.1b) (and not (O.1a)).



2.2.3.4.5) EXP occupancy

Like the two preceding ones, this profile has three parameters:

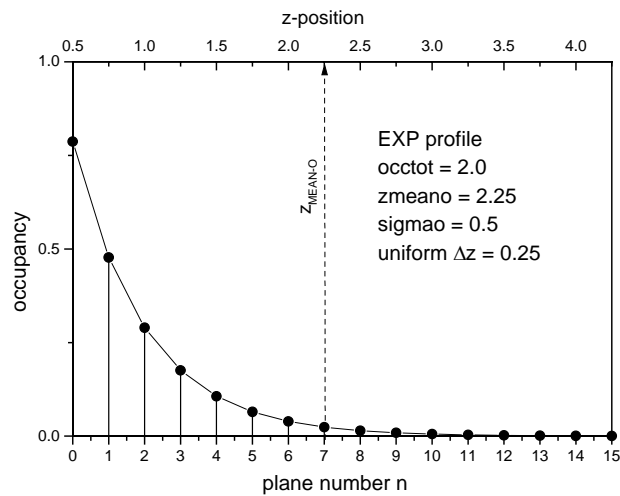
- occtot
- z_{MEANO}
- σ_{MAO} ,

that play exactly the same role as before, except for the exact form of the function.

The formula used by ROD in this case is:

$$occ(z) = occ_0 \cdot \frac{1}{2} \cdot \operatorname{erfc} \left(\frac{z - z_{MEANO}}{\sqrt{2} \cdot \sigma_{MAO}} \right) \quad (O.7)$$

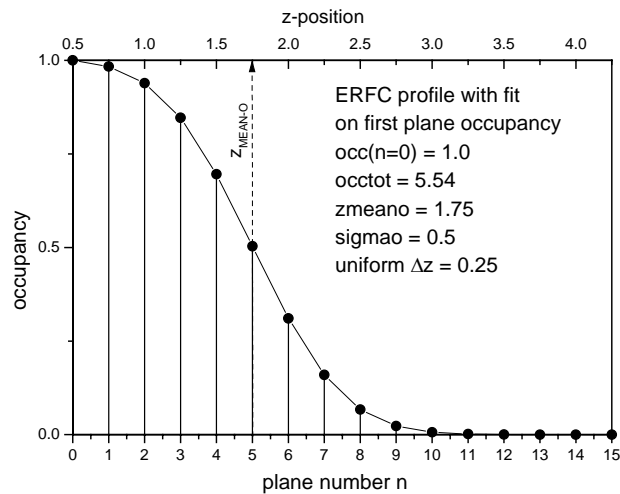
where occ_0 is derived from the condition expressed in equation (O.1a).



2.2.3.4.6) Erf occupancy function with fit on first plane occupancy

This function is exactly the same as the other erf function (see section 2.2.4.3.b) except that the condition that fixes occ_0 is not equation (O.1a) (sum of the occupancies = fixed number) but equation (O.3) (occupancy of the first plane = fixed number).

It is easier to use than the other function when you want for example to force the first adsorbate plane (s) to be completely occupied (like in the Stranski-Krastanov growth mode).



2.2.3.5) A few useful formula

2.2.3.5.1) Some considerations on roughness

Let's consider a rough surface. We number the atomic planes from $n=0$ to $n=+\infty$ (n increases when going out of the crystal), the $n=0$ plane being the last complete plane.

Let's name:

- $occ(n)$ the occupancy of plane number n
- $z(n)$ the z -position of plane number n (we choose $z(0)=0$, i.e. the origin of z is the last complete plane)
- $P(n)$ the terrace height distribution: if one takes an atomic site located at level n , $P(n)$ is the probability that this atomic site belongs to a terrace. $P(n)$ is also the fraction of the surface occupied by terraces of level n .

By considering the fact that, if an atom occupies a site at level n , it is:

- either on a terrace at level n ,
 - either in a column terminated at level $(n+1)$,
 - either in a column terminated at level $(n+2)$,
- etc...

we have:

$$occ(n) = P(n) + P(n+1) + P(n+2) + \dots \quad (R.1)$$

(we assume that there are no overhangs in the structure)

This implies:

$$occ(n+1) = P(n+1) + P(n+2) + P(n+3) + \dots \quad (R.2)$$

and so:

$$P(n) = occ(n+1) - occ(n) \quad (R.3)$$

So once we know $occ(n)$ we can easily calculate $P(n)$ and vice-versa.

How can we describe the roughness of the surface?

We can imagine forming different types of "average height", based either on the distribution $occ(n)$ or on the distribution $P(n)$. The averages that use $occ(n)$ concern all the atoms that lie above the last complete plane ($n=0$). The averages that use $P(n)$ concern only atoms that are located on terraces.

A quantity such as:

$$A_0 = \sum_{n=1}^{+\infty} occ(n) \quad (R.4)$$

will give us simply the amount of matter (in equivalent monolayers) that is present above the $n=0$ plane, i.e. the total amount of matter that form "islands" or incomplete planes.

The quantity:

$$B_0 = \sum_{n=1}^{+\infty} P(n) \quad (R.5)$$

gives simply $occ(n=1)$.

The quantity:

$$A_1 = \sum_{n=0}^{+\infty} occ(n) \cdot z(n) \quad (R.6)$$

gives the average height of the incomplete planes, with respect to the last complete plane.

The quantity:

$$B_1 = \sum_{n=0}^{+\infty} P(n) \cdot z(n) \quad (R.7)$$

gives the average height of the terraces, with respect to the last complete plane.

Another way to characterize the roughness is to take RMS values:

$$A_2 = \sqrt{\left(\sum_{n=0}^{+\infty} occ(n) \cdot (z(n) - A_1)^2 \right)} \quad (R.8)$$

which characterizes if the “islands” matter is rather concentrated around $z=A_1$ or if it is distributed over a wide z -range, and

$$B_2 = \sqrt{\left(\sum_{n=0}^{+\infty} P(n) \cdot (z(n) - B_1)^2 \right)} \quad (R.9)$$

which characterizes if the terraces are rather concentrated around $z=B_1$ or if they are distributed over a wide z -range.

B_2 is usually referred to as the “RMS roughness” of the surface.

Considering the incomplete planes of the surface as belonging to islands, one can also try to see if the shape of the islands is rather “pyramidal” or “with vertical edges” by considering how fast $occ(n)$ varies with n . Two locations where we can estimate the slope are for example: i) at the basis of the islands ($n=0$) and ii) near their middle (around $z=A_1$):

$$slope(0) = \frac{occ(n=1) + occ(n=0)}{occ(n=0) - occ(n=1)} \quad (R.10)$$

and

$$slope(N_{MEAN}) = \frac{occ(n = N_{MEAN} + 1) + occ(n = N_{MEAN})}{occ(n = N_{MEAN}) - occ(n = N_{MEAN} + 1)} \quad (R.11)$$

where N_{MEAN} is such that:

$$z(N_{MEAN}) \leq A_1 < z(N_{MEAN} + 1) \quad (R.12)$$

Those two quantities should be positive (in a model without overhangs), and the larger they are, the more “vertical” the islands edges are.

2.2.3.5.2) Diffraction by a rough surface

Let’s consider now a rough (001) surface of a crystal with a simple cubic lattice, and calculate the L dependence of the diffracted intensity on a CTR.

We assume that there is an attenuation factor α of the x-ray beam at each atomic plane, and that the attenuation starts as soon as the beam enters the crystal through a terrace.

We name:

$$x = q_z \cdot c_s \quad (R.13)$$

where q_z is the perpendicular momentum transfer ($q_z = L \cdot c_s^*$) and c_s is the lattice parameter of the crystal in the direction perpendicular to the surface.

Not taking into account the terms related to the in-plane periodicity (we take integer H and K), the structure factor of the surface is given by the coherent sum of the contributions of the different columns:

$$F_{HKL} = \sum_{n=0}^{+\infty} P(n) \cdot \sum_{j=-\infty}^{j=n} e^{i \cdot x \cdot j} \cdot \alpha^{n-j} \quad (R.14)$$

After a few manipulations, this transforms into:

$$F_{HKL} = \left(\frac{1}{1 - \alpha \cdot e^{-i \cdot x}} \right) \cdot \sum_{n=0}^{+\infty} P(n) \cdot e^{i \cdot n \cdot x} \quad (R.15)$$

Once we know the P(n) function, it is therefore very easy to calculate the roughness factor. The formulas given in section I.5) for the ERF roughness model are a direct application of formula (R.3) and (R.15).

Note that the hypothesis that we made here of a cubic crystal with only one atom per unit cell is very strong: phase shifts between different atomic planes due to x,y fractional displacements are completely forgotten.

In ROD, the formula for calculating the roughness factor uses:

$$\frac{L - L_{BRAGG}}{N_{layers}} \quad (R.16)$$

instead of L. This is a trick for dealing with more complicate crystals, like FCC (001) crystals. The thing one has to remember is that there are several cases where this trick DOES NOT work: in particular, it does not work for Si (001) surfaces.

2.2.3.5.3) Application to the “extended model”

The considerations made in section 2.3.5.1) for the roughness of a crystal can easily be extrapolated to the case of an adsorbate on a substrate.

Here in the “extended model”, we have an adsorbate with a known occupancy profile $occ(n)$ and a known $z(n)$ profile, which are directly given in the .ISL file.

Note that:

- here $z(0)$ is not necessarily equal to zero: it is the z -position of the first adsorbate plane.
- the considerations given here don't work for the Gauss occupancy profile since this profile involves overhangs.

A first useful parameter is the total amount of pseudomorphic adsorbate (in monolayers) given by:

$$occtot_{PSEUDO} = \sum_{n=0}^{+\infty} occ(n), \quad (R.17)$$

Locating the interface for example at:

$$z_{INTERF} = \frac{1}{2} \cdot (z(last_substrate_plane) + z(0)), \quad (R.18)$$

the average thickness of the pseudomorphic adsorbate is given by:

$$t_{PSEUDO} = z(0) - z_{INTERF} + \sum_{n=0}^{NPLANS-1} occ(n) \cdot (z(n) - z(0)) \quad (R.19)$$

The average height of the adsorbate terraces with respect to the origin of z is given by:

$$h_{PSEUDO} = \sum_{n=0}^{NPLANS-1} (occ(n) - occ(n+1)) \cdot z(n) \quad (R.20)$$

using the convention:

$$occ(NPLANS) = 0 \quad (R.21)$$

The RMS roughness of the adsorbate will then be:

$$\sigma_{RMS} = \sum_{n=0}^{NPLANS-1} (occ(n) - occ(n+1)) \cdot [z(n) - h_{PSEUDO}]^2 \quad (R.22)$$

The quantities given in formula (R.10) and (R.11) can be used to characterize the slope of the edges of the adsorbate islands. Note that if the adsorbate contains one or several complete planes, the basis of the islands is no more at $n=0$ but at a larger n .

2.2.3.6) The NSTART and ORIGIN parameters in the $\Delta z(z)$ functions

I never used these parameters myself so it's hard for me to guess why they have been introduced by P. Guénard. Maybe for the GAUSS occupancy? or for a model with a few complete adsorbate layers entered “by hand” and an extended model above? I don't know.

I will give the formulas anyway in case somebody finds a way to use them.

In the “non-simplified” version of ROD, where the atom ORIGIN[i] is not simply atom i, and NSTART differs from zero, for calculating the z-position of plane number n, one replaces in all the $\Delta z(n)$ and $z(n)$ formulas:

n by neff = n + NSTART
ZSFIT[i] by ZSFIT[ORIGIN[i]].

Then the atom n=0 is no more atom i but an atom located NSTART planes above the atom ORIGIN[i]. (ORIGIN[i] can be any atom number in the .fit file)

2.2.4) Listing the “extended” model: the .ISL file

The best way to check if your “extended” model behaves as you want is to go to the EXT ROB SET ADS menu in ROD and enter the command LIST. This will list either on the screen or in a file all the atoms that are in your model. The file will have an .ISL extension that stands for “full island model”.

For the non-extended atoms you get a line looking like:

i X(i) Y(i) Z(i) OCC(i)

where:

- i is the atom number in the .fit file
- X, Y, Z are the final positions of atom i in reduced coordinates
- OCC is the final occupancy of atom i.

For each extended atom i, ROD will list the NPLANS or (2 · NPLANS - 1) atoms (or layers) created by extending atom i. You get lines looking like:

i n X(n,i) Y(n,i) Z(n,i) OCC(n,i)

where:

- i is the number of the initial atom (the one that has been extended)
- n is the number of the layer (between 0 and NPLANS-1 or between -NPLANS+1 and NPLANS-1)
- X, Y, Z are the final positions of the atom of layer n
- OCC is the final occupancy of the atom of layer n.

An additional line gives you the total occupancy for the extended atom number i.

An example of an .ISL file is given in the examples section for Ge/Si(001).

2.2.5) Writing a macro file to save the options

It is possible to save into a macro file all the options of the Robach’s extension. For this you have to go to the EXT ROB menu and enter the command WRITE.

This writes all the current options in a file named ROB_EXT.MAC. After quitting the ROD program and reentering it, you can go back to the saved configuration by typing MAC ROB_EXT in the main menu (to execute the macro file).

This works better of course if you have previously read the .bul and .fit file. There are some blank lines with comments in the rob_ext.mac file that should be filled with READ BUL TOTO.BUL, READ FIT TOTO.FIT, and READ DAT TOTO.DAT type instructions.

Note that the default value of the flag for using the Robach's extension is FALSE, so you have to enter the command EXT ROB YES before you can read .bul and .fit files that are formatted for this option.

Similarly, if you want to read a data file that contains a "NLAYER" column, you have to set the "variable nlayer" flag to "yes" before you read the data file.

This is why the location of the READ commands in the macro file is not random.

Also note that by saving rob_ext.mac, you save only the options corresponding to the Robach's extension. The standard options are saved automatically in rod_init.mac and plotinit.mac when quitting the program.

2.3) Other options developed to treat the case of Ge on Si(001)

2.3.1) Introduction

The Si(001) (2x1) surface is a typical example of a "bad" case for the calculation of the Crystal Truncation Rods, because the roughness factor is particularly nasty. The key question is to know if you should make the different terraces interfere coherently with each other (which increases the size of your model more than significantly), or not. The answer is : it depends. It depends on the coherence length of your x-ray beam (the longer it is, the more you have problems), the in-plane domain size of your substrate, the mean size of the terraces, and more generally the roughness. This means that the way you model the roughness is going to depend on how rough your surface is.

The considerations developed here concerning Si (001) belong to work that is currently in progress. Which means that they can be wrong: we don't have yet a "general, simple and always applicable" model to describe the diffraction by rough Si (001).

In fact the systems where the roughness factor is easy to calculate are not so numerous: as soon as the phase shift:

$$\phi = 2 \cdot \pi \cdot (H \cdot dx + K \cdot dy + L \cdot dz)$$

between two consecutive planes is not the same for all the planes, you start having problems. Sometimes you are saved by the fact that the coherence length of the beam is smaller than the terrace size (so you can put zero roughness and do an incoherent sum of the scattering from the different terminations), but on third generation synchrotrons like ESRF this is often not the case.

It should be noted that the numerical roughness models of the standard version cannot be used to treat the problem of non-equivalent terraces, because all these models assume a constant (dx dy dz) from one plane to the other.

Some of the options we describe here are re-applicable to substrate-adsorbate systems that are much simpler than Si (001), like metals.

Some other options are more specific to systems in which, like in Si(001), different non-equivalent terraces coexist on the surface.

2.3.2) How to treat the roughness for Si(001)?

As we said in the introduction above, the calculation of the CTRs of a rough Si(001) surface is not trivial. The main question is to know if you should make the different terraces interfere *coherently* or *incoherently* with each other.

Let us first consider the case on a hypothetical clean and non-reconstructed rough Si(001) surface. This means that all the atoms in the model are at bulk-like positions.

To calculate the CTRs, one has to take into account the fact that there are two types of terraces on the Si (001): the surface can be terminated by a z=0 plane (broken

bonds along (100)_S) or by a z=1/4 plane (broken bonds along (010)_S). (Here the “S” index refers to quantities expressed in the (1x1) tetragonal unit cell).

If we calculate the CTRs using only one termination (like for an ideal surface that would be atomically flat over 1 centimeter), we find that the calculated (10L) rod is not equivalent to the (01L). In practice, when measuring the CTRs of a real Si (001) surface, the (01L) and (10L) rods are always equivalent, because of course the surface may be atomically flat over the coherent scattering length, but it is never atomically flat over centimeters so the two types of terminations necessarily coexist.

A bit of vocabulary: here we call “coherent scattering length” the smallest of the two following lengths: the in-plane domain size of the crystal near the surface (typically microns for Si), and the in-plane coherence length of the x-ray beam (typically hundreds of Å for a wiggler at LURE, thousands of Å for an undulator at ESRF).

To treat the case of a “zero roughness” Si(001) surface (i.e. a surface with terraces larger than the coherent scattering length), one already needs a model with two surface unit cells: one terminated with a (z=0)-type plane, and one terminated with a (z=1/4)-type plane.

This leads to a .fit file containing 3 atoms:

```
Si  0.5  0  0.25
Si  0.5  0.5  0.5
Si  0.5  0  0.25
```

- the first 2 atoms belong to the first surface unit cell, in which the Si is terminated with a (z=0)-type plane.
- the last atom belongs to the second surface unit cell, in which the Si is terminated with a (z=1/4)-type plane.

The theoretical intensity is obtained by adding the complex structure factor of each unit cell with the structure factor of the bulk, and then averaging the intensities of the two surface unit cells. This means an incoherent adding of the scatterings from the two terminations.

For SF=1 (SF=fraction of the surface that is reconstructed, the rest of the surface being bulk-terminated), and SF2=0.5 (fraction of the reconstructed part of the surface that is reconstructed with the second unit cell) we have:

$$I_{TOTAL} = (1/2) \cdot [|F_{BULK+F_{SURF1}}|^2 + |F_{BULK+F_{SURF2}}|^2]$$

$$= (1/2) \cdot [I_{BULK+SURF1} + I_{BULK+SURF2}]$$

The general formula used in ROD is:

$$I_{TOTAL} = (1-SF) \cdot I_{BULK} + SF \cdot [(1-SF2) \cdot I_{BULK+SURF1} + SF2 \cdot I_{BULK+SURF2}]$$

This model with two surface unit cells has a slight drawback: you are obliged to fix the “surface fraction” parameter at SF=1, otherwise you break the equilibrium between the two types of terminations.

Another simpler way to take the two terminations into account, is to take a model with 4 domains, rotated by 90° from each other. This is done by using the following commands in ROD:

```
! enter "domain" menu
set domain
! number of domains = 4
ndom 4
```

```

! to go from domain 1 to domain 1, transform (x,y) positions of
! the bulk and surface atoms using the matrix A with
! a11=1.0, a12=0.0, a21=0.0, a22=1.0 (identity matrix)
matrix 1 1.0 0.0 0.0 1.0
! domain 1 to domain 2: rotation of 90°
matrix 2 0.0 -1.0 1.0 0.0
! domain 1 to domain 3: rotation 180°
matrix 3 -1.0 0.0 0.0 -1.0
! domain 1 to domain 4: rotation 270°
matrix 4 0.0 1.0 -1.0 0.0
! equal occupancy for all the domains
equal YES

```

For the case of only one surface unit cell, ROD uses the formulas:

$$I_{\text{BULK}} = \sum \text{DOMOCCUP}[j] \cdot I_{\text{BULK}}[j]$$

$$I_{\text{SURF}} = \sum \text{DOMOCCUP}[j] \cdot I_{\text{SURF}}[j]$$

$$I_{\text{BULK+SURF}} = \sum \text{DOMOCCUP}[j] \cdot (|F_{\text{BULK}}[j] + F_{\text{SURF}}[j]|^2)$$

$$I_{\text{TOTAL}} = (1-\text{SF}) \cdot I_{\text{BULK}} + \text{SF} \cdot I_{\text{BULK+SURF}}$$

where the sum Σ is over the different domains “j”, and DOMOCCUP[j] is the occupancy of domain number j (here 1/4 for all the domains).

The Si (001) crystal is not invariant by a 90°-rotation in the surface plane, but it is invariant by the combination of a 90°-rotation and a shift of 1/4 in the z-direction. Since by adding the intensities of the different domains, we lose the phase information associated to the relative shift between the different domains, putting 4 domains terminated at z=0 is equivalent (in ROD) to using 4 surface unit cells located at z=0, 1/4, 1/2 and 3/4.

In contrast with the model “two surface unit cells + 1 domain”, the model “1 surface unit cell + 4 domains” allows to vary the “surface fraction” parameter without breaking the equilibrium between the different terminations. It is also significantly simpler to modify when one starts adding a reconstruction.

Let us now discuss the case of a rough non-reconstructed Si (001) surface, i.e. a surface with terraces smaller than the coherent scattering length. As for the “zero-roughness” surface, we need to consider two surface unit cells (or 4 domains), to account for the fact that the last complete plane (occ=1) may be either a z=0 plane or a z=1/4 plane. But now we also need to consider the possibility of coherent adding of the scatterings from the two types of terraces.

For bulk-terminated Si, the trick of using $(L-L_{\text{BRAGG}})/\text{NLAYERS}$ in the roughness factor works for rods such as (20L) and (11L) for which the Bragg peaks are separated by $\Delta L=4$, but not for the rods such as (10L) and (30L), for which the Bragg peaks are separated by $\Delta L=2$. The $(L-L_{\text{BRAGG}})/\text{NLAYERS}$ trick does not work for the (10L) rod because the phase shift $d\phi=2\pi \cdot (H \cdot dx + K \cdot dy + L \cdot dz)$ from one plane to the other is not constant.

An approximate way of dealing with the roughness for a bulk-terminated Si (001) surface consists in using the $(L-L_{\text{BRAGG}})/\text{NLAYERS}$ trick in the roughness factor, but with a variable NLAYERS: one uses NLAYERS=4 for the (20L) and (11L) rods, and NLAYERS=2 for the (10L) and (30L) rods.

This model led to introduce in the program an option that allows different NLAYERS for the different data points. This option is presented in the next section.

We checked that this model gives a reasonable agreement with our data taken at ESRF on a clean Si (001) (2x1) surface (10L, 20L, 30L, and 11L rods measured up to L=4).

We also compared the results of this approximate calculation with the results of a complete calculation of the Si(001) CTRs for the case of a β^n roughness model.

The exact expression for the structure factor at (HKL) is:

$$F0(\alpha, H, K, L) := 1 + \alpha \cdot e^{2\pi i \cdot \left(\frac{K-L}{2}\right)} + \alpha^2 \cdot e^{2\pi i \cdot \left(\frac{H+K-L}{2}\right)} + \alpha^3 \cdot e^{2\pi i \cdot \left(\frac{H-3L}{2}\right)}$$

$$F1(\alpha, H, K, L) := e^{2\pi i \cdot \left(\frac{H+L}{2}\right)} + \alpha + \alpha^2 \cdot e^{2\pi i \cdot \left(\frac{K-L}{2}\right)} + \alpha^3 \cdot e^{2\pi i \cdot \left(\frac{H+K-L}{2}\right)}$$

$$F2(\alpha, H, K, L) := e^{2\pi i \cdot \left(\frac{H+K+L}{2}\right)} + \alpha \cdot e^{2\pi i \cdot \left(\frac{H+L}{2}\right)} + \alpha^2 + \alpha^3 \cdot e^{2\pi i \cdot \left(\frac{K-L}{2}\right)}$$

$$F3(\alpha, H, K, L) := e^{2\pi i \cdot \left(\frac{K+3L}{2}\right)} + \alpha \cdot e^{2\pi i \cdot \left(\frac{H+K+L}{2}\right)} + \alpha^2 \cdot e^{2\pi i \cdot \left(\frac{H+L}{2}\right)} + \alpha^3$$

$$F(\alpha, \beta, H, K, L) := \frac{1-\beta}{1-\alpha^4 \cdot e^{-2\pi i \cdot L}} \cdot \frac{1}{1-\beta^4 \cdot e^{2\pi i \cdot L}} \cdot \left(F0(\alpha, H, K, L) + \beta \cdot F1(\alpha, H, K, L) + \beta^2 \cdot F2(\alpha, H, K, L) + \beta^3 \cdot F3(\alpha, H, K, L) \right)$$

$$i := 0..600 \quad L_1 := \frac{i}{100} \quad \text{atten} := 0.0001 \quad \alpha := e^{-\frac{\text{atten}}{4}} \quad \alpha = 1 \quad \beta := 0.2$$

$$I10L_1 := \left(|F(\alpha, \beta, 1, 0, L_1)| \right)^2 \quad I01L_1 := \left(|F(\alpha, \beta, 0, 1, L_1)| \right)^2$$

$$I_{ave}10L_1 := \frac{1}{2} \cdot (I10L_1 + I01L_1)$$

where α is the attenuation factor per plane (typically, atten=0.0001 in ROD and $\alpha = \exp(-\text{atten}/4)$). Here the atomic scattering factor and Debye-Waller factor have been omitted for simplicity.

The formula used in ROD is:

$$\text{Frod}(\alpha_{\text{rod}}, \beta_{\text{rod}}, H, K, L, L_{\text{bragg}}, n_{\text{layers}}) := \left| \frac{1 - \beta_{\text{rod}}}{1 - \beta_{\text{rod}} \cdot e^{-2 \cdot \pi \cdot i \cdot \frac{L - L_{\text{bragg}}}{n_{\text{layers}}}}} \right| \cdot \frac{\text{FO}(1, H, K, L)}{1 - \alpha_{\text{rod}} \cdot e^{-2 \cdot \pi \cdot i \cdot L}}$$

$$cs := 5.43 \quad n11 := 4 \quad n12 := 2 \quad \sigma_{\text{rms}} := \frac{cs \sqrt{\beta}}{n11 \cdot (1 - \beta)} \quad dum := \left(\frac{cs}{n12 \cdot \sigma_{\text{rms}}} \right)^2$$

$$\beta_{\text{rod1}} := \beta \quad \beta_{\text{rod2}} := \left(\frac{1}{2} \right) \cdot \left[2 + dum - \sqrt{dum \cdot (dum + 4)} \right] \quad \alpha_{\text{rod}} := \alpha^4$$

$$\beta_{\text{rod1}} = 0.2 \quad \beta_{\text{rod2}} = 0.068 \quad \sigma_{\text{rms}} = 0.759$$

$$I_{\text{rod10L}_i} := \left(\left| \text{Frod}(\alpha_{\text{rod}}, \beta_{\text{rod2}}, 1, 0, L_i, 1, 2) \right| \right)^2$$

$$I_{\text{rod01L}_i} := \left(\left| \text{Frod}(\alpha_{\text{rod}}, \beta_{\text{rod2}}, 0, 1, L_i, 1, 2) \right| \right)^2$$

$$I_{\text{rodave10L}_i} := \frac{1}{2} \cdot (I_{\text{rod10L}_i} + I_{\text{rod01L}_i})$$

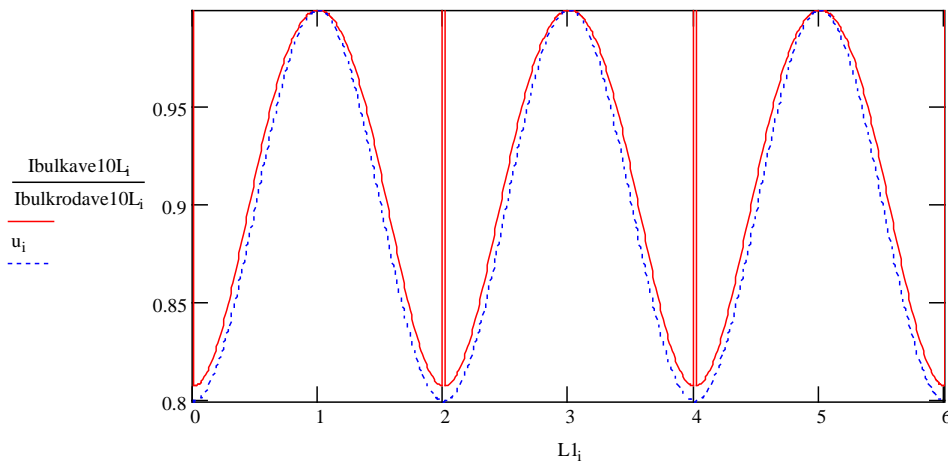
where α_{rod} is the attenuation factor per unit cell used in ROD ($\alpha_{\text{rod}} = \alpha^4$). β_{rod} is equal to $\beta_{\text{rod1}} = \beta$ for the 20L rod, and to β_{rod2} for the (10L) rod. n_{layers} is equal to 2 for the (10L) rod and to 4 for the (20L) rod. β_{rod2} is deduced from β_{rod1} by the condition that the rms roughness σ_{rms} , which depends on both β and n_{layers} , should be the same for the two rods. L_{bragg} is equal to 1 for the (10L) rod, and 0 for the (20L) rod.

To take into account the two terminations, one needs to take the average of the intensities of the (HKL) and (-KHL) rods.

The numerical calculation shows that the calculation used in ROD is a rather good approximation of the complete calculation. The ratio ($I_{\text{AVE-10L}} / I_{\text{ROD-AVE-10L}}$) is between 0.9 and 1.0 for $\beta = 0.1$, and between 0.5 and 1.0 for $\beta = 0.9$. This ratio has a rather simple shape as a function of L , and can be described approximately by the formula:

$$\frac{I_{\text{AVE-10L}}}{I_{\text{ROD-AVE-10L}}} = 1 - \frac{1}{2} \cdot \sin\left(\frac{\pi}{2} \cdot \beta\right) \cdot \cos^2\left(\frac{\pi}{2} \cdot L\right)$$

$$u_i := 0.9 - 0.1 \cdot \cos(\pi \cdot L_i)$$



It should be noted that this is valid only after averaging the intensities of the (10L) and (01L) rods. Before averaging, the differences between the two calculations are

much larger. In particular, the approximate calculation done in ROD tends to exaggerate notably the difference between the (01L) and (10L) rods, compared to the exact calculation. At $\beta=0$, the two calculations give identical results. When β increases, the difference between the (10L) and (01L) rods starts to decrease for the exact calculation, while it remains large for the approximate calculation.

Here we used $\text{atten}=0.0001$ in all the calculations. A larger attenuation factor for the incident beam may change the results.

Let us now move on to the case of a rough Si (001) (2x1) surface. The structure of the (2x1) reconstruction of Si(001) is well-known. The surface is terminated by asymmetric dimers of Si, which form rows perpendicular to the dimers. The calculation of the roughness factor becomes heavier in this case because the $z=0$ terraces are reconstructed (2x1) (dimer axis along x) and the $z=1/4$ terraces are reconstructed (1x2) (dimer axis along y).

Again, one has to put either 2 unit cells or 4 domains in the model, to account for the two terminations.

In the limit of $\alpha=1$ (no attenuation of the incident beam) and for a β^n roughness model, the contribution of the reconstructed planes to the complex structure factor is given by:

$$\text{Frec}(\beta, H, K, L) := \frac{(1-\beta)}{1-\beta^4 \cdot e^{2\pi \cdot 1i \cdot L}} \cdot \left[\begin{array}{l} \text{Fsurf}(H, K, L) \dots \\ + \beta \cdot e^{2\pi \cdot 1i \cdot \left(\frac{H+L}{2} + \frac{L}{4}\right)} \cdot \text{Fsurf}(K, -H, L) \dots \\ + \beta^2 \cdot e^{2\pi \cdot 1i \cdot \left(\frac{H+K+L}{2}\right)} \cdot \text{Fsurf}(-H, -K, L) \dots \\ + \beta^3 \cdot e^{2\pi \cdot 1i \cdot \left(\frac{K}{2} + \frac{3L}{4}\right)} \cdot \text{Fsurf}(-K, H, L) \end{array} \right]$$

where

$$\text{Fsurf}(H, K, L) = \sum_j \text{occ}(j) \cdot \exp(2 \cdot i \cdot \pi \cdot (H \cdot x_j + K \cdot y_j + L \cdot z_j))$$

is the standard expression for the contribution of the surface unit cell, the sum over j being a sum over all the atoms in the surface unit cell. The surface unit cell may as usual extend over several planes. Here we assume that the last complete plane of the bulk contains the atom at (0 0 0).

This complex structure factor Frec needs to be added to the contribution of the bulk:

$$\text{Ftot}(\alpha, \beta, H, K, L) := \text{F}(\alpha, \beta, H, K, L) + \text{Frec}(\beta, H, K, L)$$

and then the intensity is averaged over the two terminations.

$$\text{Itot}_{10L_i} := \left(\left| \text{Ftot}(\alpha, \beta, 1, 0, L_i) \right| \right)^2 \quad \text{Itot}_{01L_i} := \left(\left| \text{Ftot}(\alpha, \beta, 0, 1, L_i) \right| \right)^2$$

$$\text{Itotave}_{10L_i} := \frac{1}{2} \cdot (\text{Itot}_{10L_i} + \text{Itot}_{01L_i})$$

The approximate expression used in ROD is:

$$\text{Frecrod}(\beta_{\text{rod}}, H, K, L, \text{Lbragg}, \text{nlayers}) := \left| \frac{1 - \beta_{\text{rod}}}{1 - \beta_{\text{rod}} \cdot e^{2\pi \cdot 1i \cdot \frac{L - \text{Lbragg}}{\text{nlayers}}}} \right| \cdot \text{Fsurf}(H, K, L)$$

Here we used as an example the model of the Si (001) (2x1) reconstruction given by R. Felici et al (Surf. Sci. 375 (1997) p 55). The numerical calculation shows that the relative intensities of the different rods are rather well described by the approximate calculation by ROD, except for the 20L rod. In the exact calculation, the (20L) shows a bump near $L=2$ that is not present in the approximate calculation. This bump is clearly due to the coherent interference between the y-axis-dimers of one terrace and the x-axis-dimers of the terrace below. When β increases, this bump increases in amplitude and also becomes narrower. At $\beta=0.9$, it has a shape resembling a small Bragg peak at $L=2$.

Such a bump was present in our data, with a very low amplitude for Si(001) (2x1), and with a larger amplitude for Sb/Si (001), which means that this effect has to be taken into account. This effect cannot be reproduced with a zero-roughness surface, except if one allows both x and y displacements of the atoms of the model (twisted dimers).

The 11L rod is identical in the two calculations. On the (11L) and the (30L) rods, the ratio between the two calculations (I_{tot}/I_{totrod}) shows an oscillatory behavior as a function of L, being equal to 1 at the Bragg peaks, and to 0.25 to 0.5 near the anti-Bragg points for $\beta=0.9$. The bump in the (20L) rod near $L=2$ corresponds to a ratio of 1.8 between the two calculations for $\beta=0.2$, and a ratio of 275 for $\beta=0.9$. Again, the discrepancy between the two calculations increases with β , for all the rods except the (11L).

This comparison shows that the calculation currently used in ROD is unable to reproduce some of the characteristic oscillations present in the data. It also shows that the coherent interference between non-equivalent terraces can add some non-trivial oscillatory behavior in the truncation rods. This kind of oscillations does not bring any additional information about the atomic positions: it just tells us that there is some “coherent roughness” on the surface.

These oscillations may be less well-defined in reality than in the calculation. The relative sizes of two consecutive terraces can vary from one domain to the other, which changes the ratio between the x- and y-dimers, and this is likely to smear the oscillations. This is not related to which occupancy profile is used for describing the roughness: we are just saying that, even if the β^n profile gives a good average description of the surface, the real surface may deviate locally from this ideal profile. This is true for any rough surface, but usually we neglect these effects since the roughness causes only a general decrease of the intensity at all L's. In special cases like Si(001), where roughness can cause locally an increase of the intensity, we need to consider this eventuality.

2.3.3) The VARIABLE NLAYERS option

As was mentioned above, for bulk-terminated Si (001), a good approximate way to describe the roughness is to use the (L-LBragg)/Nlayers trick with Nlayers=4 for the (20L) and (11L) rods, and Nlayers=2 for the (10L) and (30L) rods.

Since the rms roughness σ of the surface depends on both β and Nlayers (see section I.5), if we change Nlayers we need to change β accordingly to keep the same σ for all the rods.

With this new option, one can add another column “nlayer” to the data file, so that different values of Nlayers are used for the different (H K L) data points. This works only with the “approximated beta” roughness model. When the flag for the “variable nlayers” is set to yes, we use SIGMA (the rms roughness) instead of BETA as the fitting parameter. For each (H K L), we calculate from Nlayers and SIGMA the value of β that should be used, using the formula given in section I.5 for the β^n roughness model.

In practice the choice of the “variable Nlayers” mode is done in the ROD.EXT.ROB.SET.PAR menu, by entering the command “VAR YES”. In this menu, one can also set the value of SIGMA.

2.3.4) The **RATIO** option

This option can be useful in any growth study. We developed it for solving the following problem.

We had a data set of three truncation rods of Si (001) measured on the clean Si (001) and for several deposits of Ge. The problem was that the alignment was not perfect (although it was imperfect in the same way during all the experiment), so the fit on the clean Si was far from ideal. As a consequence, when we started adding pseudomorphic Ge in the model, all the fit was doing was adding Ge to compensate the initial bad fit of the Si, instead of adding Ge that would account for the DIFFERENCES that appeared in the Si CTRs when adding the Ge.

In this case, the difference between the rods with and without Ge was much more relevant than the absolute intensity of the rods with the Ge. So we added into ROD the “RATIO” option, which allows to do a fit on the quantity:

$$\frac{|F(\text{adsorbate}+\text{substrate})|}{|F(\text{clean substrate})|} \quad (\text{here } F(\text{Si+Ge})/F(\text{clean Si}))$$

instead of directly

$$|F(\text{adsorbate}+\text{substrate})|.$$

This option gives less weight to the high intensity points close to the Bragg peaks, and more weight to the low intensity points, where most of the oscillatory behavior appears: what drives the fit is how much the rod has changed at one point, not how close the point is to the Bragg peak.

In ROD, you can select the “RATIO” option by entering the menu EXT ROB SET ADS CALC and typing the command RATIO. This asks you which mode you want to work in, the ratio mode (data = F(ads+sub)/F(sub)) or the normal mode (data = F(ads+sub)).

By default, ROD assumes that all the atoms in the .FIT file (“extended” or “non-extended”) are ADSORBATE atoms, and that all the atoms in the .BUL file are SUBSTRATE atoms. (in version 1.1 one had to give ROD the numbers of the adsorbate atoms otherwise the ratio was always 1)

2.3.5) variation of the interface roughness (when using the **RATIO** option)

This option is a sub-option of the RATIO option. As the RATIO option, it can be useful for any growth study. It was introduced to treat the following problem.

In the event of an interdiffusion between the Ge and the Si, we had to be able to take into account the fact that the roughness of the Si could vary when we deposited the Ge (this way of treating the interdiffusion is in fact rather crude). This is why, for the ratio mode, we introduced a second roughness parameter BETA2, to describe the roughness of the substrate before the deposition. In short, the theoretical ratio is:

$$\frac{|F(\text{adsorbate}+\text{substrate}) * f_{\text{rough}}(\text{BETA})|}{(F(\text{substrate}) * f_{\text{rough}}(\text{BETA2}))}$$

where f_{rough} is the attenuation factor due to the roughness (formula (11) in E. Vlieg’s “A concise ROD manual”). BETA is a fitting parameter, but BETA2 is not: it is fixed at the value of BETA obtained on the clean substrate.

2.3.6) The **ATOM** option: what to do when the .BUL file is not enough to describe the clean substrate (when using the **RATIO** option)

This option is again a sub-option of the RATIO option.

In its present form, it allows to define in the .fit file which atoms are SUBSTRATE atoms (by default all the atoms of the .FIT file are considered as ADSORBATE atoms). The idea is to put the occupancy of the ADSORBATE atoms to zero when we calculate $F(\text{substrate})$ for the ratio $F(\text{substrate} + \text{adsorbate}) / F(\text{substrate})$.

This option is only useful if the substrate has a reconstruction initially, and if the deposition of the adsorbate does not change the atomic positions of the substrate (this is not the case for Ge on Si (001) (2x1), because the Ge destroys the Si reconstruction).

The setting of which atoms of the .fit file are SUBSTRATE atoms is done in the menu EXT ROB SET ADS CALC, by entering the command ATOM. This asks you first how many atoms in the .fit file are SUBSTRATE atoms, and then what are their numbers in the .fit file.

In the future, we plan to add to the RATIO option the possibility of using two completely different surface unit cells for calculating $F(\text{substrate})$ and $F(\text{adsorbate} + \text{substrate})$. This would enable to use the RATIO option in the case where the clean substrate is reconstructed and the deposition of the adsorbate lifts the substrate's reconstruction.

2.3.7) The ZSHIFT parameter

The ZSHIFT(i) parameter enables to have a different origin in the occupancy function for each extended atom i: this replaces $\text{occ}(z)$ by $\text{occ}(z - \text{ZSHIFT}(i))$.

Originally, this enabled to solve the following problem:

For Ge on Si(001), we have two surface unit cells, one with the Si terminated at $z=0.5$, and one with the Si terminated at $z=0.25$. Let's say that we use directly z and not $(z - \text{ZSHIFT})$ in the occ function, and that we take the same occupancy function for the two surface unit cells. In this case, the first Ge plane of the first unit cell will not have the same occupancy as the one of the second unit cell, since it is at $z=0.75$ instead of $z=0.5$. This causes a problem because it breaks the equilibrium between the two types of terminations and renders the (01L) rod non-equivalent to the (10L) rod.

The good variable to consider in the occ function is not simply z , but the quantity:
[$z - z(\text{last substrate plane})$]

which leads to introducing a parameter $\text{ZSHIFT} = z(\text{last substrate plane})$.

This option is in fact not useful for Ge / Si(001) because one can take 4 domains instead of 2 surface unit cells to describe the clean bulk-terminated Si.

2.3.8) Further developments planned

We plan to add something to treat the case where the adsorbate grows in islands and there is a reconstruction at the surface of the adsorbate. This will be done for fitting data taken on deposits of Ge on Si (001) grown in presence of a surfactant (Sb).

The implementation should not be very difficult: we need to create a "reconstruction unit cell" with an occupancy profile along z that is proportional to the size of the terraces of the adsorbate islands. For the case of a layer of Sb floating at the top of the Ge islands, this means taking an occupancy profile $\text{occSb}(z)$ for the Sb equal to the first derivative of the occupancy profile of the Ge, $\text{occGe}(z)$.

$$\text{occSb}(z) = d(\text{occGe}(z))/dz$$

or

$$\text{occSb}(\text{level } n) = \text{occGe}(\text{level } n-1) - \text{occGe}(\text{level } n)$$

We also need to link the (x,y,z) positions of the Sb atoms at level n+1 to the (x,y,z) positions of the Ge atoms located at level n.

Another planned development is to introduce “complementary” occupancy profiles, of the form (1-occ(z)), which would enable to treat interdiffusion at interfaces and segregation using the extended model. This should be fairly straightforward.

2.4) Extension of the “PG - extended model” to RELAXED adsorbates

2.4.1) Introduction

In sections 2.1) to 2.3), we wrote about the interferences and oscillations that can appear on the substrate CTRs when we deposit an adsorbate. In those sections, we assumed that the interferences come from PSEUDOMORPHIC adsorbate. This is not always true: they can also come from RELAXED adsorbate.

Ag/MgO(001) and Ni/Ag (001) are examples of this latter situation: Ag/MgO has a low misfit (3%) but a very low adhesion energy, so pseudomorphic Ag is unlikely; Ni/Ag(001) has a very large misfit (14% for the cube on cube epitaxy) so pseudomorphic Ni is also unlikely. Interference however appears on the substrate CTRs in both systems.

A bit of terminology. We call *pseudomorphic* the adsorbate that has the same in-plane lattice parameter as the substrate, i.e. the adsorbate that is strained by the substrate, and *relaxed* the adsorbate that has recovered at least partially its bulk in-plane lattice parameter. Here we mean by “partially” that the adsorbate can have an in-plane lattice parameter intermediate between the one of bulk adsorbate and the one of the substrate. We do not mean that one part of the adsorbate is at its bulk lattice parameter and that the rest is pseudomorphic.

This idea that relaxed adsorbate can modify the CTRs of the substrate is at first thought a bit strange, since relaxed adsorbate gives Bragg peaks that are at a different value of the in-plane momentum transfer q_{\parallel} than the substrate CTRs. We do not expect intuitively to have any interference between two contributions that are located at different places in the reciprocal space. In addition, we expect the scatterings from the different adsorbate domains to add incoherently since there is no particular phase relationship between the domains.

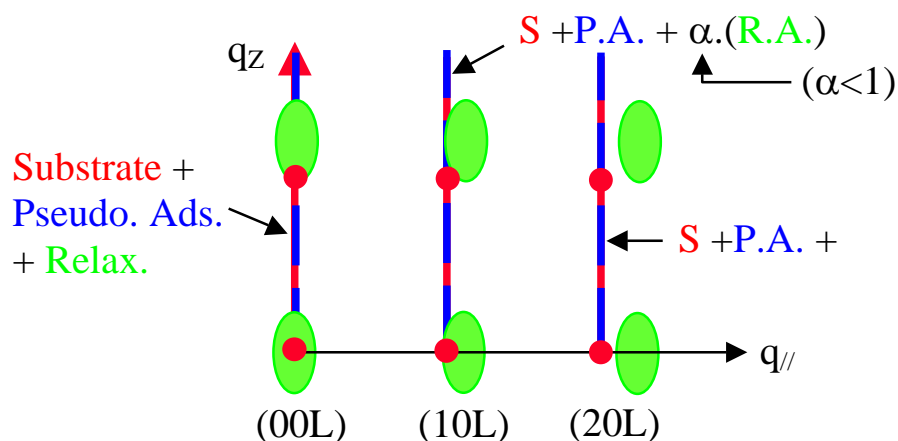
This simple picture is in fact not always true, for the following reason. Imagine that we are depositing the adsorbate on the substrate from a vapor, in a way that is more or less “atom by atom”. In the very first stages of the deposit, the adsorbate atoms will be dispersed on the surface of the substrate, as “one atom” islands. If there is a preferential adsorption site (for example on top of oxygen atoms for Ag on MgO) they will adsorb on it without any question (we assume that there are no “low energy” adsorption sites due to defects like steps or vacancies). Imagine also that the adsorbate has a tendency to form relaxed islands on this particular substrate. Then the atoms arriving next on the surface will tend to gather around the “seed” atoms that are already there and form islands. The initial “seed” atom at the center of the island will tend to keep its low energy adsorption site, but the atoms arrived later will have “seats” that are not so good, because of the lattice mismatch and the tendency of the adsorbate to relax.

The idea is that, even if the adsorbate already starts to relax, since there is a “seed” atom, the region around the center of the islands will tend to have a constant structure throughout the deposit : in this region the positions of the adsorbate atoms with respect to the substrate will be the same for all the islands. This remains true even if differences appear between the islands regarding their size, their shape or their internal strain field, or in the event the islands coalesce : in this latter case the columns

of atoms correlated via the substrate that were initially at the center of the islands will remain in the continuous layer (this was observed for Ni/Ag).

The presence of “seed atoms” therefore introduces “repeating units” in the deposit, that have the very important property of being separated by lattice vectors of the substrate. The existence of those “repeating units” in the adsorbate is at the origin of the interference on the substrate CTRs. Indeed, at the location of the substrate CTRs, and at this location only, the scatterings from to the different repeating units add *coherently*, not only with each other but also with the scattering from the substrate.

What we will get in reciprocal space is described in the figure below: this is a drawing of the reciprocal space seen in a H-L plane (H: direction parallel to the surface, L: direction perpendicular to the surface), for a generic adsorbate-substrate system. Here we assume that the two materials are cubic and in cube-on-cube epitaxy, and that the adsorbate has a smaller lattice parameter than the substrate.



There are three sources of scattering that can contribute to the CTRs of the substrate:

- first the substrate itself (in red),
- secondly the pseudomorphic adsorbate (in blue), whose rods are at the same H,K positions as the ones of the substrate,
- and thirdly the relaxed adsorbate (in green), whose rods are shifted in H,K with respect to the ones of the substrate, and which can contribute to the CTRs of the substrate if there is a correlation (via the substrate) between the domains of relaxed adsorbate.

If we consider the specular (00L) rod, all the sources contribute with the same weight. This rod is not sensitive to the lateral crystallographic order in the deposit.

For non-zero H, K values, the substrate and the pseudomorphic adsorbate always contribute with the same weight, but the weight of the relaxed adsorbate decreases, with a greater and greater decrease as H and K increase.

A good test for knowing if a deposit contains pseudomorphic adsorbate is therefore to measure a substrate CTR with big values of H and K, and to see if this CTR gets modified when one deposits the adsorbate. For example, for Ag/MgO (001) and Ni/Ag(001), it was clearly established that all the interference observed on the substrate CTRs come from relaxed adsorbate : there is no pseudomorphic adsorbate.

2.4.2) The modeling of the CTRs intensity

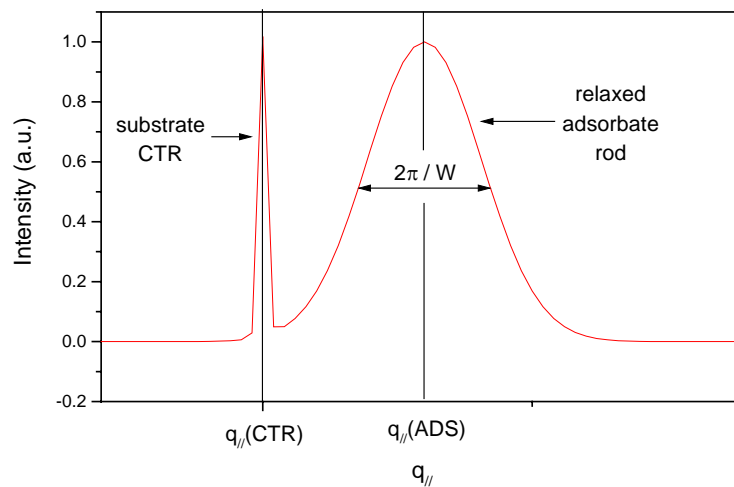
How can we model that with a mathematical form simple enough to be used in ROD?

Of course we cannot start with a complete description of the deposit, involving both the morphology of the islands and their internal strain field, because that would mean many more parameters than we can handle.

Without going into so many details, it is relatively easy to foresee the main differences between interferences due to pseudomorphic adsorbate and interferences due to relaxed adsorbate.

If we take a pseudomorphic adsorbate, the rods of the adsorbate are located at the same $q_{//}$ as the rods of the substrate, for all the possible values of $q_{//}$. Therefore, the relative weight of the contributions of the adsorbate and the substrate to the truncation rods will not vary with $q_{//}$ (except for a possible difference in the Debye Waller factor between the two materials). If we take now a relaxed adsorbate, the rods of the adsorbate are not located at the same $q_{//}$ as the rods of the substrate, and the spacing between the adsorbate and substrate rods increases with increasing $q_{//}$. Therefore, when $q_{//}$ increases, the contribution of the adsorbate to the substrate CTRs will decrease with respect to the contribution of the substrate.

In the drawing below, we have represented what could be a radial in-plane scan through a substrate CTR and a relaxed adsorbate rod. The diffracted intensity is plotted versus $q_{//}$. The substrate CTR, centered on $q_{//\text{CTR}}$, is very sharp due to the large in-plane domain size of the substrate (We work only with good crystals, don't we?). The adsorbate rod, centered on $q_{//\text{ADS}}$, is much broader, due to the small size of the adsorbate islands.



The contribution of the adsorbate to the substrate CTR comes from the intensity that remains in the tail of the adsorbate peak at $q_{//}=q_{//\text{CTR}}$. Since the width of the adsorbate peak is proportional to the inverse of the width W of the adsorbate domains (we neglect the lattice parameter distribution), the adsorbate contribution to the substrate CTR will contain a Debye-Waller-like factor involving the width W and the misfit f , namely :

$$DW_{RELAX}(W, f) = \exp \left[-\frac{\|\vec{q}_{//CTR} - \vec{q}_{//ADS}\|^2}{\left(\frac{2 \cdot \pi}{W}\right)^2} \cdot C \right] \quad (A.1)$$

$$DW_{RELAX}(W, f) = \exp \left[-\|\vec{q}_{//CTR}\|^2 \cdot f^2 \cdot \left(\frac{W}{2 \cdot \pi}\right)^2 \cdot C \right]$$

where f is the in-plane misfit between the substrate and the adsorbate (we take cubic materials in cube on cube epitaxy to simplify):

$$f = \frac{a_{ADS} - a_{SUB}}{a_{SUB}} \quad (A.2)$$

and

$$C = 2 \cdot Ln(2) \quad (A.3)$$

Here we assumed that the adsorbate peak has a Gaussian intensity profile in $q_{//}$ with a full-width-at-half-maximum (FWHM) equal to $(2 \cdot \pi / W)$. This gives a Gaussian profile with a FWHM in $q_{//}$ of $(2\sqrt{2} \cdot \pi / W)$ for the modulus of the structure factor.

The Gaussian shape of the peak is of course rather arbitrary.

To transform a model of pseudomorphic adsorbate into a model of relaxed adsorbate in ROD, all one has to do is to enter the value of the misfit, to add the width of the adsorbate domains to the list of parameters, and to multiply the adsorbate contribution to the substrate CTRs by the DW_{RELAX} factor described above. The rest of the “PG –extended model” is absolutely the same as in sections 2.2 and 2.3.

Since for islands small enough to have no interfacial defects, the island size is a good approximation of the domain size, the fits of the substrate CTRs can in principle (and rather surprisingly) be used to determine the islands lateral size. Since the parameters of the $occ(z)$ function in the extended model give an estimate of the height of the adsorbate islands, the fit of the substrate CTRs can provide (although indirectly) some information on the morphology of a deposit.

Once we know the $occ(z)$ profile and the islands width W , we can also get an idea of the inter-islands distance, d_i .

Assuming for a rough estimate that we have islands with a square basis arranged on a square lattice, we have:

$$occ(n) = \frac{(W(n))^2}{d_i^2} \quad (A.4)$$

where $W(n)$ is the width of the islands at level n . If we assume that the islands have a circular basis and are arranged on a hexagonal lattice, this formula becomes:

$$occ(n) = \frac{\pi}{2 \cdot \sqrt{3}} \cdot \frac{(W(n))^2}{d_i^2} \quad (A.5)$$

which is not so much different.

Knowing that:

$$W = \frac{1}{N_{occ}} \cdot \sum_{n=0}^{NPLANS-1} W(n) \quad (A.6)$$

where N_{occ} is the number of planes for which $occ(n)$ is non-zero, we can deduce d_i by the formula:

$$d_i = W \cdot \frac{N_{occ}}{\sum_{n=0}^{NPLANS-1} \sqrt{occ(n)}} \quad (A.7)$$

This formula has to be used with caution, since the $occ(z)$ profiles used in the program have sometimes very long tails with a very small value of $occ(n)$ over a large number of planes, so there is a risk of overestimating N_{occ} and therefore d_i . A reasonable choice of N_{occ} is such that, if n is between 0 and N_{occ} , the value of $W(n)$ calculated from d_i and $occ(n)$ should be at least larger than one interatomic distance. (this is a kind of self-consistent calculation).

Of course all of this is oversimplified:

- domains sizes are not always equal to islands sizes
 - the shape of the adsorbate peak is not necessarily a Gaussian,
 - crystals are not always cubic, and they do not always grow in cube on cube epitaxy,
- but all in all this formalism has the merit of being relatively simple.

2.4.3) Implementation in ROD

To introduce relaxed adsorbate in the model for fitting the CTRs, we need to know:

- what is the crystalline structure of the adsorbate?
 - is it a bulk structure, and if so in which phase?
 - is it an epitaxy-stabilized non-bulk structure?
 - is only one structure present or is there a mixture of several structures?
- what is the epitaxial relationship between the adsorbate and the substrate?
- what is the in-plane lattice parameter of the adsorbate, and is there an anisotropy of this parameter between the different in-plane directions?

To answer all those questions, one needs to do additional surface-x-ray diffraction measurements, such as:

- looking for adsorbate peaks at the places where they are expected for the different possible epitaxial relationships
- locating them precisely in H and K to get the in-plane lattice parameters
- measuring their in-plane width in different directions to get the in-plane domain size, the mosaic and the lattice parameter distribution.

For the moment the program can only deal with adsorbate / substrate systems that have a square in-plane unit cell, and that are in “cube-on-cube” epitaxy. The choice between a pseudomorphic adsorbate and a relaxed adsorbate is done in the menu EXT ROB SET ADS CALC, by entering the command RELAX. This asks you which mode (relaxed or pseudomorphic) you want to work in. Then in the MISFIT menu you can enter the value of the misfit (as defined in eq. (A.2)). In the WIDTH menu you can enter the width (in Å). This width will afterwards be used as a fitting parameter.

For Ag/MgO(001), the lines to add to the .MAC file are typically:

```
ext rob set ads calc
relax yes
misfit -0.03
width 50 0.00 0.00 NO
return return
```

A few additional remarks:

- normally the domain width derived from the fit of the CTRs should be about the same as the one deduced from the in-plane scans on the adsorbate peaks.
- note that the total amount of relaxed adsorbate derived from the fit of the CTRs is usually smaller than the deposited amount: only the fraction of the adsorbate that is correlated via the substrate contributes to the substrate CTRs.
- in the .ISL file, the atomic positions of the “extended” atoms will always be listed as if the adsorbate was pseudomorphic, even if the calculation of the rods has been done with a hypothesis of relaxed adsorbate.
- in the fits, there is a very strong coupling between the “occtot” and “width” parameters so up to now we had to fix at least one of them to avoid getting an error in the fits.
- this option has just been implemented so there may be bugs in it.
- when calculating the “surface” part of the structure factor, the multiplication by the $DW_{RELAX}(W,f)$ factor applies only to the scattering by the “extended” atoms. The scattering by the “non-extended” atoms of the fit file stays the same as usual.
- here we considered only the two extreme cases: pseudomorphic adsorbate or relaxed adsorbate. Of course one can also imagine a deposit where the two kinds of adsorbate coexist: for example in Pt/Ni we see partially relaxed Pt from the beginning of the deposition, but some of the Pt of the first layer seems to be intermixed with the Ni, giving a signal corresponding to “pseudomorphic” Pt on the Ni CTRs. The treatment of this kind of situation is not yet implemented in ROD.

3) Examples of ROD input files

Here are the .bul, .fit, and .mac files that we used in ROD for the fits of:

- Ag/MgO (001) (fit of $F(Ag+MgO)$)
- Ge/Si (001) (more complicated, fit of $F(Ge+Si)/F(clean\ Si)$)

The example files are listed together with a description of the contents of the various columns for the .bul and .fit files.

3.1) Ag/MgO(001)

3.1.1) The .BUL file

```
mgo.bul
mgo bulk
```

```

4.212 4.212 4.212 90 90 90
O      0      0      0      1      0
O      0.5    0.5    0      1      0
Mg     0.5    0      0      1      0
Mg     0      0.5    0      1      0
Mg     0      0      -0.5  1      0
Mg     0.5    0.5    -0.5  1      0
O      0.5    0      -0.5  1      0
O      0      0.5    -0.5  1      0

```

first line: comments

second line: direct lattice parameters (lengths + angles):

as bs cs α s β s γ s

other lines: list of the bulk atoms:

Element X Y Z SN(DW) SN(OCC)

where

- X, Y, Z are the positions of the atom in reduced coordinates
- SN(DW) is the serial number of the Debye-Waller parameter
- SN(OCC) is the serial number of the occupancy parameter

The z-axis is pointing outward the crystal. The topmost bulk plane should be at $z=0$.

3.1.2) The .FIT file

a) Standard format

agl.fit

```

1 ML Ag strained (Ag above the O atoms of mgo.bul)
4.212 4.212 4.212 90 90 90
Ag 0.0 0.0 0 0.0 0 0.0 0.0 0 0.0 0 0.5 1 1 0 0 2 0 1 0
Ag 0.5 0.0 0 0.0 0 0.5 0.0 0 0.0 0 0.5 1 1 0 0 2 0 1 0

```

first line: comments

second line: direct lattice parameters: (they must be the same as in the .bul file)

as bs cs α s β s γ s

other lines: list of the surface atoms:

```

Element XS XCONST NXDIS X2CONST NX2DIS
YS YCONST NYDIS Y2CONST NY2DIS
ZS ZCONST NZDIS Z2CONST NZ2DIS
NDWS NDWS2
NOCCUP NOCCUP2

```

where

- XS, YS, ZS are the starting positions of the atom in reduced coordinates
- NXDIS, NYDIS and NZDIS are the serial numbers of the first displacement parameter along the x, y and z directions respectively
- XCONST, YCONST and ZCONST are the multiplication factors of the first displacement along the x, y and z directions respectively
- NX2DIS, NY2DIS and NZ2DIS are the serial numbers of the second displacement parameter along the x, y and z directions respectively
- XCONST, YCONST and ZCONST are the multiplication factors of the second displacement along the x, y and z directions respectively

- NDWS, NDWS2 are the serial number of the Debye-Waller parameter (parallel and perpendicular to the surface)
- NOCCUP is the serial number of the occupancy parameter
- NOCCUP2 is the serial number of the 2nd occupancy parameter (optional)

To explain how all those things work:

- for the meaning of the displacement and occupancy parameters: see section I.6
- for the meaning of the occupancy and Debye-Waller parameters: it is easier to simply give the corresponding code lines (from the source file calc.c):

```
/* for each atom i of the surface unit cell */
if (NDWS2[i] != 0)
    fatom = fat[TS[i]] * dw_par[NDWS[i]] * dw_perp[NDWS2[i]];
else
    fatom = fat[TS[i]] * dw[NDWS[i]];
if (NOCCUP[i] != 0) fatom *= OCCUPTOT[NOCCUP[i] - 1];
```

where

```
dw_par[j] = exp(-DEBWAL[j-1]*q_par*q_par/PIPI16);
dw_perp[j] = exp(-DEBWAL2[j-1]*q_perp*q_perp/PIPI16);
dw[j] = exp(-DEBWAL[j-1]*q*q/PIPI16);
fat[i] = f_atomic(q/2,i);
```

where DEBWAL and DEBWAL2 are the Debye-Waller factors on which we do the fitting, q_{par} and q_{perp} are the parallel and perpendicular components of the momentum transfer, q is the modulus of the momentum transfer and PIPI16=16.π². f_{atomic} is a function that calculates the atomic scattering factor using the value of q and coefficients that depend on the element. These coefficients can be found in the international tables for x-ray crystallography.

```
f = F_COEFF[natom][8];
for (i = 0; i < 4; i++)
{
    f += F_COEFF[natom][i*2]*
        exp(-F_COEFF[natom][i*2+1]*sqr(0.5*q_half/PI));
}
```

Note that the displacements serial numbers go from 1 to N, including the three displacements directions x, y and z.

When using two surface unit cells :

The first (NSURFTOT-NSURF2) atoms of the fit file belong to the first unit cell, while the last NSURF2 atoms of the fit file belong to the second unit cell where:

- NSURF2 is specified in the SET CALC menu
- NSURFTOT is the total number of atoms in the .fit file

b) MCSL's format

When the MCSL extension is activated, one can use an alternative format for the .FIT file, which allows for more flexibility: only the useful columns need to be put in

the .FIT file, and the order of the columns does not matter as long as it matches the order of the column names given in the 3rd line of the file.

See the manual for MCSL's extension for more details.

Below is a .FIT file that is equivalent to ag1.fit but with the new format.

ag1short.fit

```
1 MC Ag constraint (Ag au-dessus des atomes d'O de mgo.bul)
4.212 4.212 4.212 90 90 90
#el XS YS ZS cz1 nz1 dw1 ocl
Ag 0 0 0.5 1 1 2 1
Ag 0.5 0.5 0.5 1 1 2 1
```

To use this format, the MCSL extension needs to be activated before reading the .FIT file, using the command EXT MCSL ACT YES.

3.1.3) The .MAC file

With this version of ROD, the nesting of macros files is authorized. Therefore, instead of writing a .PAR file and then reading successively the .BUL, .FIT, .DAT and .PAR files, one can create a single .MAC file that does all the work in one command. To execute a macro file called toto.mac, one has to enter the command MAC TOTO in the main menu. Lines starting by an exclamation mark “!” are interpreted as comments.

```
agmgo.mac

! Go to extensions robach menu
ext rob
! activate Robach's extension
activate YES
! go back to main menu
return return

! Read files .bul, .fit and .dat
read bulk mgo.bul
read fit ag1.fit
read data 03mc.dat

! Go to set calc menu - standard parameters
set calc
! data = structure factors (not intensities)
struct yes
! attenuation coefficient of incident beam with z
atten 0.0001
! nlayers, used in roughness calculation
nl 2
! number of atoms in the second surface unit cell
nsurf2 0
return return
! go to set parameter menu
set par
! sets the various parameters
! scale factor of theory
scale 1.00 0.00 0.00 NO
! roughness parameter beta
beta 0.57 0.00 0.00 NO
! fraction of the surface that is reconstructed
surfrac 1.00 0.00 0.00 NO
! displacement with serial number 1
displace 1 0.10 0.00 0.15 YES
```

```

! Debye-Waller parameter with serial number 1
b1      1   0.30   0.00   0.00  NO
! idem with serial number 2
b1      2   0.66   0.00   0.00  NO
! occupancy parameter with serial number 1
occ     1   0.05   0.00   1.00  NO
return return
! go to set domains menu
set domain
! number of domains
ndom 1
! do not interpret non-integer H,K as fractional orders
frac no
! equal occupancy for the different domains = no
eq no
! occupancy of domain 1 = 1.0
occ 1 1.0
return return

! Go to extensions robach set par menu
ext rob set par
! use erf roughness model
erfrough YES
! set Lshift between exp and theory to 0.0
lshift 0.0000 0.0000 0.0000 NO
return
! Go to extensions robach set fit menu
fit
fullmodel yes
return
! Go to extensions robach set ads extended menu
ads
! set atom 1 as an extended atom
extended 1
mode yes
! for this atom, use dz(z) profile number 1
zdis 1
! for this atom, use maximum number of plane of 20
nplans 20
! for this atom, the repeating unit along z contains 2 planes
nlayer 2
! for this atom, the 2nd plane of the repeating unit is shifted
! in-plane by dx = 0.5 with respect to the first plane
xshift 2 0.5
! for this atom, use occ(z) profile number 1
odis 1
! for this atom, the origin of the occ(z) profile is atom 1
origin 1
return
! set atom 2 as an extended atom
extended 2
mode y
! etc ...
zdis 1
nplans 20
nlayer 2
xshift 2 -0.5
odis 1
origin 2
return return
! Go to ext robach set ads func menu
function
! dz functions:
! add an dz(z) function with serial number 1
zadd 1
! sets the parameters for the dz(z) function number 1
! type = 0, uniform dz=dz0
! dz0 = 0.485, with a range checking between 0.43 and 0.50

```

```

zpar 1 0 0.485 0.43 0.50 YES
! occ functions:
! add an occ(z) function with serial number 1
oadd 1
! sets the parameters for the occ(z) function number 1
! type = 1, erf function with fit on total occupancy
! center of erf function at zmean=0.5, with range checking
! width of erf function = dzrough = 0.5, with range checking
opar 1 1 0.5 0.5 10 YES 0.5 0.0 10.0 YES
return
! Go to ext robach set ads calc menu
calc
! use normal calculation : data = |F(adsorbate + substrate)|
ratio no
! adsorbate is relaxed
relax yes
! in-plane domain size of adsorbate domains = 20 angstroms
width 20 0.00 0.00 NO
! misfit between adsorbate and substrate is -3 percent
misfit -0.03
return return return return return

```

See E. Vlieg's "ROD manual" for the details, as well as the preceding sections of this manual.

Note for DW factors: they are entered as rms displacements values of thermal vibrations $\langle u_x^2 \rangle^{1/2}$ (units = Å).

Default values are (when serial number = 0) :

- zero for the displacements
- one for the occupancies
- zero for the Debye-Waller factors

3.2) Ge / Si (001)

3.2.1) The .BUL file

si6.bul

```

Si (001)(1x1) si6.bul
3.8400 3.8400 5.4306 90.0 90.0 90.0
Si 0.50 0.50 0.00 1 0
Si 0.00 0.50 -0.25 1 0
Si 0.00 0.00 -0.50 1 0
Si 0.50 0.00 -0.75 1 0

```

3.2.2) The .FIT file

ge.fit

```

Ge on Si (001) surface
3.8400 3.8400 5.4306 90.0 90.0 90.0
Ge 0.5 0 0 0 0 0.0 0 0 0 0 0.25 1 0 0 0 2 0 0

```

3.2.3) The .MAC file

```
sige.mac
```

```

! version of 08-11-00. 4 domains, variable nlayers
! ratio mode, Ge/Si(001), no reconstruction

```

```

! Go to extensions robach menu
ext rob
activate YES
return return
! read .bul and .fit files (model)
read bulk si6.bul
read fit ge.fit
! Go to set calc menu - standard parameters
set calc
struct yes
atten 0.001
sfrac 1.0
scale 3.39
rough approx
return return
set par
bl 1 0.2 0.00 0.00 NO
bl 2 4.35 0.00 0.00 NO
disp 1 0.012 0.00 0.00 NO
occ 1 2.67 0.00 0.00 NO
return return
! 4 domains to take the different terminations into account
set domain
ndom 4
matrix 1 1.0 0.0 0.0 1.0
matrix 2 0.0 -1.0 1.0 0.0
matrix 3 -1.0 0.0 0.0 -1.0
matrix 4 0.0 1.0 -1.0 0.0
equal YES
frac no
return return

! Go to extensions robach set par menu
ext rob set par
erfrough NO
lshift 0.0000 0.0000 0.0000 NO
! use variable nlayers from data file
var yes
! set rms roughness sigma to 5.44 angstroms
sig 5.44 0.000 0.000 NO
return
! Go to extensions robach set fit menu
fit
fullmodel yes
! Go to extensions robach set ads extended menu
return
ads
extended 1
mode y
zdis 1
nplans 20
nlayer 4
zshift 0.0
xshift 2 -0.5
xshift 3 -0.5
yshift 3 0.5
yshift 4 0.5
odis 1
origin 1
return
! Go to ext robach set ads func menu
function
! dz functions:
zadd 1
zpar 1 0 0.26 0.20 0.30 YES
! occ functions:
oadd 1
opar 1 1 0.5 0.25 6.0 YES 0.1 0.0 6.0 YES

```

```

return
! Go to ext robach set ads calc menu
calc
! use ratio mode: data= F(ads+sub)/F(sub)
ratio yes
! the .fit file contains no substrate atoms
atom 0
! the adsorbate is pseudomorphic
relax no
! set rms roughness sigma for clean substrate to 3.0 angstroms
sig 3.000
return return return return
! read data file
read data rge9minb.dat

```

3.2.4) The .ISL file

Here is the .ISL file that ROD creates when using the macro file sige.mac, and then the command EXT ROB SET ADS LIS.

```

Atom El  Plane   X    Y    Z    Occ
first surface unit cell :
1   Ge   0    0.5000  0.0000  0.2620  1.8817
1   Ge   1    0.0000  0.0000  0.5220  0.7838
1   Ge   2    0.0000  0.5000  0.7820  0.0046
1   Ge   3    0.5000  0.5000  1.0420  0.0000
1   Ge   4    0.5000  0.0000  1.3020  0.0000
1   Ge   5    0.0000  0.0000  1.5620  0.0000
1   Ge   6    0.0000  0.5000  1.8220  0.0000
1   Ge   7    0.5000  0.5000  2.0820  0.0000
1   Ge   8    0.5000  0.0000  2.3420  0.0000
1   Ge   9    0.0000  0.0000  2.6020  0.0000
1   Ge  10    0.0000  0.5000  2.8620  0.0000
1   Ge  11    0.5000  0.5000  3.1220  0.0000
1   Ge  12    0.5000  0.0000  3.3820  0.0000
1   Ge  13    0.0000  0.0000  3.6420  0.0000
1   Ge  14    0.0000  0.5000  3.9020  0.0000
1   Ge  15    0.5000  0.5000  4.1620  0.0000
1   Ge  16    0.5000  0.0000  4.4220  0.0000
1   Ge  17    0.0000  0.0000  4.6820  0.0000
1   Ge  18    0.0000  0.5000  4.9420  0.0000
1   Ge  19    0.5000  0.5000  5.2020  0.0000
Total occupancy for atom 1: 2.6700
extended atom 1:
ofunction SN 1 type=erfc occupancy with fit on total occ
zfunction SN 1 type=no relaxation

```

4) In practice

Here is a description of the ROD menus corresponding to the Robach's extension, as they appear on the display in the program.

```

ROD>he
***** MAIN MENU *****
Read      : Read data/model files
List      : List data/model
RESet     : Reset all parameters
Calculate : Calculate structure factors
Plot      : Goto plotting menu
Set       : Set parameters

```

```

Fit      : Fit experimental structure factors
ENergy   : Goto lattice energy menu
EXtensions: Goto extensions menu
Macro    : Run macro file
:        : Execute an operating system command
Help     : Display menu
QUIT     : Quit program
ROD>ext
ROD.EXT>he
***** EXTENSIONS *****
Robach   : go to Robach's extensions menu
SVensson : go to Svensson's extensions menu
Walker   : go to Walker's extensions menu
MCsl     : go to MCSL's extensions menu
List     : list current extension
Help     : Display menu
Return   : Return to last menu
ROD.EXT>ro
ROD.EXT.ROB>he
***** ROBACH MENU *****
Activate : Activate/deactivate Robach's extension
Set      : Set features of Robach's extension
Write    : Write current features to macro file
RESet    : Reset all param of this extension
Help     : Display menu
Return   : Return to main menu
ROD.EXT.ROB>ac
Activate Robach's extensions ? [YES]:
ROD.EXT.ROB>set
ROD.EXT.ROB.SET>he
***** ROBACH SET MENU *****
Parameters: Parameters for rod calculation
Fit       : Model par. for fitting surface structure
ADsorbate : Parameters describing the adsorbate
SLab      : Slab model, used for binary alloys
ANomalous : Corr. of at. scatter. factor near edges
Help      : Display menu
Return    : Return to main menu
ROD.EXT.ROB.SET>par
ROD.EXT.ROB.SET.PAR>he
***** ROBACH SET PARAMETERS *****
Erfrough  : Use approx erf model for roughness
LShift    : Lshift between exp and theory
SC3ale    : Scale factor for frac orders
S2urffrac : Fraction of rec. surf. with 2nd unit cell
Varnlayer : Use variable NLAYERS from data file
Sigma     : Rms roughness - only for variable NLAYERS
Fsurfrough: Apply bulk roughness to fsurf
List      : List parameters
Help      : Display menu
Return    : Return to main menu
ROD.EXT.ROB.SET.PAR>
ROD.EXT.ROB.SET.PAR>lis
Use approximated erf model for roughness: NO
scale factor of fractional orders: 0.0000 [ 0.0000, 0.0000] NO
Lshift: 0.0000 [ 0.0000, 0.0000] NO
frac. of rec. surface rec. with 2nd unit cell: 0.0000 [ 0.0000,
0.0000] NO
Use variable NLAYERS from data file.
rms roughness sigma : 5.4400 [ 0.0000, 0.0000] NO
ROD.EXT.ROB.SET.PAR>
ROD.EXT.ROB.SET>fit
ROD.EXT.ROB.SET.FIT>he
***** ROBACH SET FIT MENU *****
Element   : Element type of atom
Xstart    : Start x-position
XConstant : Mult. factor of x-displacement
XDisplace : Serial # of x-displacement parameter

```

```

X2Constant: Mult. factor of 2nd x-displ.
X2Displace: Serial # of 2nd x-displacement par.
Ystart      : Start y-position
YConstant   : Mult. factor of y-displacement
YDisplace   : Serial # of y-displacement parameter
Y2Constant: Mult. factor of 2nd y-displ.
Y2Displace: Serial # of 2nd y-displacement par.
Zstart      : Start z-position
ZConstant   : Mult. factor of z-displacement
ZDisplace   : Serial # of z-displacement parameter
Z2Constant: Mult. factor of 2nd z-displ.
Z2Displace: Serial # of 2nd z-displacement par.
B1          : Serial # of in-plane Debye-Waller par
B2          : Serial # of out-of-plane D-W par
Occupancy   : Serial # of occupancy parameter
O2occupancy: Serial # of 2nd occ. parameter
Add         : Add an atom to model
Delete      : Delete an atom from model
Fullmodel   : Show extended model yes/no
List        : List atoms
Help        : Display menu
Return     : Return to main menu
ROD.EXT.ROB.SET.FIT>lis
# el xs xconst nxdis x2const nx2dis ys yconst nydis y2const ny2dis zs
zconst nzdis z2const nz2dis ndw ndw2 nocc nocc2
 1 Ge 0.5000 0.00 0 0.00 0 0.0000 0.00 0 0.00 0
0.2500 1.00 1 0.00 0 2 0 1 0
ROD.EXT.ROB.SET.FIT>
ROD.EXT.ROB.SET>ads
ROD.EXT.ROB.SET.ADS>he
***** ROBACH SET ADSORBATE MENU *****
Extended  : Parameters for extending surface atoms
Functions : Occ(z) and dz(z) function parameters
Calc      : Options for extended model calculation
List      : List atoms of extended model
Help      : Display menu
Return    : Return to main menu
ROD.EXT.ROB.SET.ADS>lis
Filename (.isl) (type 't' or <return> for terminal):
Atom      El      Plane  X      Y      Z      Occ
first surface unit cell :
1         Ge      0       0.5000 0.0000 0.2620 1.8817
1         Ge      1       0.0000 0.0000 0.5220 0.7838
1         Ge      2       0.0000 0.5000 0.7820 0.0046
1         Ge      3       0.5000 0.5000 1.0420 0.0000
1         Ge      4       0.5000 0.0000 1.3020 0.0000
1         Ge      5       0.0000 0.0000 1.5620 0.0000
1         Ge      6       0.0000 0.5000 1.8220 0.0000
1         Ge      7       0.5000 0.5000 2.0820 0.0000
1         Ge      8       0.5000 0.0000 2.3420 0.0000
1         Ge      9       0.0000 0.0000 2.6020 0.0000
1         Ge     10      0.0000 0.5000 2.8620 0.0000
1         Ge     11      0.5000 0.5000 3.1220 0.0000
1         Ge     12      0.5000 0.0000 3.3820 0.0000
1         Ge     13      0.0000 0.0000 3.6420 0.0000
1         Ge     14      0.0000 0.5000 3.9020 0.0000
1         Ge     15      0.5000 0.5000 4.1620 0.0000
1         Ge     16      0.5000 0.0000 4.4220 0.0000
1         Ge     17      0.0000 0.0000 4.6820 0.0000
1         Ge     18      0.0000 0.5000 4.9420 0.0000
1         Ge     19      0.5000 0.5000 5.2020 0.0000
Total occupancy for atom 1: 2.6700
extended atom 1 :
ofunction SN 1 type=erfc occupancy with fit on total occ
zfunction SN 1 type=no relaxation
ROD.EXT.ROB.SET.ADS>
ROD.EXT.ROB.SET.ADS>ext
Atom number [return]: 1

```



```

ROD.EXT.ROB.SET.ADS.EXT>he
***** ROBACH SET ADS EXTENDED *****
Mode      : Extended or standard calculation mode
ZDis      : SN of dz distr. function
NStart    : Initial atom is in plane nb NSTART
NPlans    : Total z extension (in nb of planes)
NLayers   : Number of planes within cell height
Xshift    : X trans. between planes within cell
Yshift    : Y trans. between planes within cell
ODis      : SN of occ distr. function
ORigin    : Z of 1st plane = Z orig + NSTART*dz
ZShift    : Shift to subtract from zm in o.func.
List      : List parameters
Help      : Display menu
Return    : Return to previous menu
ROD.EXT.ROB.SET.ADS.EXT>lis
Atom 1 parameters
extended mode
z function: 1
atom belonging to plane: 0
max number of planes: 20
number of inequivalent planes in cell: 4
    x shift for layer 1: 0.0000
    y shift for layer 1: 0.0000
    x shift for layer 2: -0.5000
    y shift for layer 2: 0.0000
    x shift for layer 3: -0.5000
    y shift for layer 3: 0.5000
    x shift for layer 4: 0.0000
    y shift for layer 4: 0.5000
occupancy function: 1
shift to subtract from zm in occ func: 0.0000
origin atom number: 1
ROD.EXT.ROB.SET.ADS.EXT>
ROD.EXT.ROB.SET.ADS>func
ROD.EXT.ROB.SET.ADS.FUNC>he
***** SET ADS FUNCTIONS *****
ZAdd      : Add interplanar distance dist. z func
OAdd      : Add occupancy distribution z function
ZDel      : Remove interpl. dist. dist. z func.
ODel      : Remove occup. distribution o function
ZPar      : Set z function parameter
OPar      : Set o function parameter
List      : List parameters
Help      : Display menu
Return    : Return to previous menu
ROD.EXT.ROB.SET.ADS.FUNC>lis
z functions:
1: no relaxation          dz(z->+oo) : 0.2600 [0.2000,0.3000] YES
o functions:
1: erfc occupancy with fit on total occ
    zmean: 0.5000 [0.2500,6.0000] YES
    sigma: 0.1000 [0.0000,6.0000] YES
ROD.EXT.ROB.SET.ADS.FUNC>
ROD.EXT.ROB.SET.ADS>calc
ROD.EXT.ROB.SET.ADS.CALC>he
***** ROBACH SET ADS CALC MENU *****
Ratiomode : Set calc. mode: ratio / normal
B2eta     : In ratio mode: value of beta for Fsub
AToms     : In ratio mode: indicate substrate atoms
RELaxmode : Set calc. mode: pseudomorphic / relaxed
Width     : In relaxed mode: width of ads. domains
Misfit    : In relaxed mode: misfit (ads-sub)/sub
Sigma2    : In ratio + var. nl mode: sigma for Fsub
List      : List parameters
Help      : Display menu
REturn    : Return to main menu

```

```

ROD.EXT.ROB.SET.ADS.CALC>lis
Use ratio Fads+sub over Fsub (ratio mode)
Use variable NLAYERS.
rms roughness sigma used for Fsub = 3.0000
Number of SUBSTRATE atoms in surface unit cell = 0
adsorbate is PSEUDOMORPHIC
ROD.EXT.ROB.SET.ADS.CALC>

```

5) Some bugs

- Under linux: one needs to put the “.bul” and “.fit” extensions in non-caps letters in the file names. If the bulk file has a .BUL extension or any other extension, one needs to give the complete file name when reading the file in ROD (idem for the “fit”, “sur”, and “dat” files).
- In the menu SET DOMAIN: when using only one domain (command NDOM 1), the command EQUAL YES tends to give strange things. That’s why we usually put EQUAL NO and then we have to enter the occupancy of domain 1 using the command (OCC 1 1.0) (otherwise the default is zero).
- In this version when calculating a rod and then doing a PLOT ALL (to plot Fbulk, Fsurf and Ftot versus L) the Fbulk and Fsurf curves are dashed lines and the Ftot curves is a solid line.
- When using a data file in which H and K are very close to integers but not exactly (typically 1.999), one has to do SET DOMAIN FRAC NO command (otherwise the data points are interpreted as being fractional orders and the calculation gets messed up).
- When doing a calculation with only the .BUL file (no .FIT file), i.e. with only the bulk unit cell, if by any chance SURF2FRAC (the fraction of the reconstructed surface occupied by the second surface unit cell) is different from zero, the calculation tends to get messed up. It is better to do a SET CALC S2 0.0 to avoid that.
- Problems with plotting menus:

There are two plotting menus in ROD, a first one in the main menu (ROD.PLOT>) that enables to plot data, fits and models, and a second one (PLOT>) that enables to do some plot manipulations (overlays etc...) and to change the settings for the plots.

To access the second menu (PLOT>) one needs to plot at least one curve, for example by the command PLOT DATA in the main menu. After hitting the ENTER key for this command one arrives in the PLOT> menu. Another hitting of the ENTER key brings you back to the ROD> menu.

Here is one bug we noticed: when the errors bars are activated (command PLOT ER YES from ROD>), one cannot anymore access the second plot menu (PLOT>). One needs to type the command PLOT ER NO in ROD> before one can access again the PLOT> menu.

- When one plots experimental and theoretical structure factors stored in a .LST file, if ROD has just been re-opened, one needs to read a .BUL file first, otherwise the program crashes with a floating-point error when doing the plot command. (the .LST file is normally obtained with a LIST COMPARE command). This is true for all the plotting commands that need to draw the in-plane unit cell, because they need the values of the lattice parameters.

6) Some questions

- Isotropic / anisotropic Debye-Waller factors:

In the .FIT file, we have two columns, one for the serial number of the parallel (//) Debye-Waller (DW) factor, and one for the serial number of the perpendicular (\perp) DW factor. This allows anisotropic DW factors. In case one wants an isotropic DW factor, there are two solutions: either use the same serial number for SNDW (DW //) and SNDW2 (DW \perp), or more simply use a certain serial number for SNDW (DW //), and put zero for SNDW2 (DW \perp). ROD interprets SNDW2=0 as meaning that the DW is isotropic, with a serial number given by SNDW.