REFTIM 7_4

MANUAL

GENERAL

The program REFTIM calculates and fits the time spectra of reflectivity from any multilayer structure containing ^{57}Fe, ^{151}Eu, ^{119}Sn or ^{149}Sm resonant nuclei[1] for given grazing angles. The calculation and fit of the prompt and delayed reflectivity curves for the same model of the structure is also included. You are able to compare the theoretical spectra with the experimental ones on the graphs and to improve the suggested model of the multilayer structure, HFI parameters and depth distribution function for each hyperfine field by handle or automatic variation of these parameters. The initial comparison of the initial and new structure model is mostly effective "by eyes" on the time spectrum graphs and after that a refinement could be done sequentially by "one-parameter" descending with the chosen MERIT function. You also can keep an eye on the obtaining variations of the spectrum graphs "on line". You also can watch graphs of the initial and next energy spectra of absorption and reflectivity (button "**Energy graphs**") as well as the electronic and nuclear HFI density depth distribution (button "**Model graphs**") for the used model in dialog windows.

We use the interface step functions not only for the electronic density and photoabsorption (i.e. for the real and imaginary parts of the electronic susceptibility) but also for each kind of the hyperfine interaction separately. Five interface functions (linear, arctangent, half Gauss, Gauss and Error function) can be automatically depicted by steps on the scale of the assigned interface thickness for calculations of reflectivity The interface width and the number of steps should be the same for all used interface functions for the layer.

In the last version REFTIM 7.4 we have inserted the field distribution functions.

The essential improvement of the REFTIM program is the possibility to fit the set of the time spectra simultaneously with the delayed and prompt curves. This option is very essential for many tasks such as the delayed Bragg peak diminish by diffusion in periodic multilayers, standing wave enhancement of the delayed reflectivity peaks etc.

The program has been created by Marina Andreeva (marina@trtk.ru or Mandreeva1@yandex.ru) with the essential corrections by Bengt Lindgren (Bengt.Lindgren@fysik.uu.se) and the very effective practical help by Vitalii Panchuk (vitpan@mail.ru).

The kind request

If you discover some mistakes, instability, misprints or other unclear moments in the program or this program description **please inform us** *by*
*Mandreeva1@yandex.ru*

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[1]The program works for any M1 transitions in Mössbauer nuclei if the special data file "**Tconst.INI**" with specific nuclear parameters is prepared – see the description below.

OPERATIONS

At first you should unpack "**REFTIM_pack.ZIP**" to some chosen directory. (attention! The program does not like the names of directories with blank spaces). The package contains "**REFTIM.exe**", "**TIM09F.exe**", "**DEL09F.exe**", "**REF09F.exe**" and "**CEM09F.exe**" files and some data files with description of the needed properties for 4 Mossbauer transitions 14.4 keV in ^{57}Fe, 21.532 keV in ^{151}Eu, 23.871 keV in ^{119}Sn and 22.494 keV in ^{149}Sm: "**FeConst.INI**, "**EuConst.INI**", "**SnConst.INI**", "**SmConst.INI**". The file "**Tconst.INI**" is used during the calculations and it is reloaded, when you choose the needed isotope by "**Settings**" → "**Select isotope**" from the menu-line.

The program has started by "**REFTIM.exe**". The main window is opened.
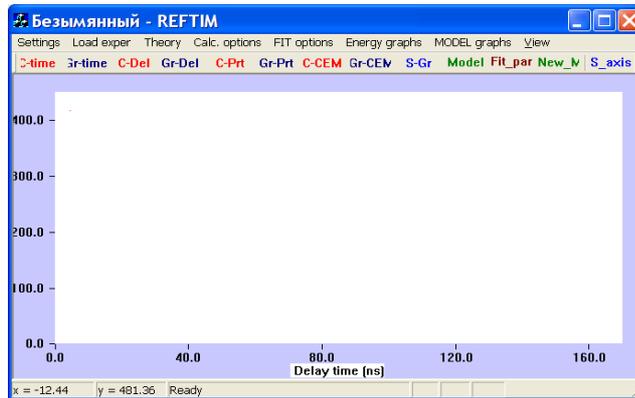
It is recommended also to press **"Settings"** → **"Clean previous result",** in such a way you delete the temporary files with the previous calculations in the working directory.

If you have worked with the data previously you can simply choose **"Settings"** → **"Get previous settings"**. The program will accept the information about the experimental data in consideration and the calculation options used in previous attempts. In this case it is supposed that the service files: "**AXISF.INP**", "**FILEM.INP**", "**FILESE.INP**", "**NEXT.INP**", "**STOP.INP**", "**TIM09F.INP**", "**DEL09F.INP**", "**FilCEM.INP**" and "**Tconst.INI**" have already existed in the chosen directory, as well as the used model file (like "**m.inf**" in the attached "**example**" directory) and files with the experimental data (in the attached "**example**" directory they are "**Dat_Tim.DAT**", "**Dat_Del.DAT**", "**CEMS.DAT**").

It you work with the program for the first time you press **"Settings"** → **"Start from zero"** and all needed service files: "**AXISF.INP**", "**FILEM.INP**", "**FILESE.INP**", "**NEXT.INP**", "**STOP.INP**", "**TIM09F.INP**", "**DEL09F.INP**", "**FilCEM.INP**" are created automatically.

When you LOAD EXPERIMENT (by the button "**Load Exper**" or by the button **"Settings"** → **"Start from zero"**) you should point out the amount Nspc and the names of the data files with experimental time spectra (for different grazing angles but for one and the same sample so the model should be the same for all the time spectra, delayed and prompt curves or CEM spectrum). In default the number of the experimental spectra is 0 and the program can work just with theoretical calculations. If Nspc≠0, the graphs of the experimental time spectra immediately appear on the screen. In the same dialog you should put the names of the files with delayed and prompt curve and CEMS spectrum (if you have them).
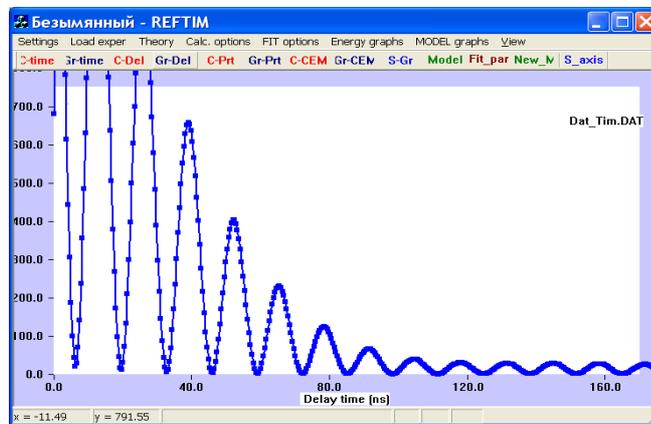
The experimental data file preparation

Notice that the number of points in the experimental time spectra should not be greater than 900. We use the sum over 5 points for ESRF experimental 2048 point spectra and get 330 point spectra, in ORIGIN that is the following transformation of the columns:

(col(A)[(i-1)*5+1]+col(A)[(i-1)*5+2]+col(A)[(i-1)*5+3]+col(A)[(i-1)*5+4]+col(A)[(i-1)*5+5])/5
(col(B)[(i-1)*5+1]+col(B)[(i-1)*5+2]+col(B)[(i-1)*5+3]+col(B)[(i-1)*5+4]+col(B)[(i-1)*5+5])

We keep the backward sequence of the time scale of the experimental and sometimes slightly shift the time scale according to the shape and the position of the prompt pulse. And we extract the needed time range (10 –170 ns). So our example data files for analysis contain only about ~ 300 points. The example experimental data file is "**Dat_Tim.DAT**" in the attached "**example**" directory – the first column is the delay time (ns), the second column is the corresponding experimental reflectivity count. If the program finds the pointed files the experimental time spectra (or several spectra) appear in the window.

For the delayed and prompt reflectivity the example of the experimental data file is "**Dat_Del.DAT**" in the attached "**example**" directory: the first line is the number of points in the measured angular dependence, the first column is the glancing angle θ (in mrad !), the second column is the prompt reflectivity count, the third column is the delayed reflectivity count. The name of the delayed and prompt reflectivity experimental data file can be pointed out also in the same dialog window "**Load Experiment**" or in the service file "**DEL09F.INP**", which can be reached and corrected e.g. by the buttons "**Calc. options**" →"**Del and prompt**". The experimental curves (normalized) should appear on the window if you press **"Gr-Del" or "Gr-Prt"** hot-keys.
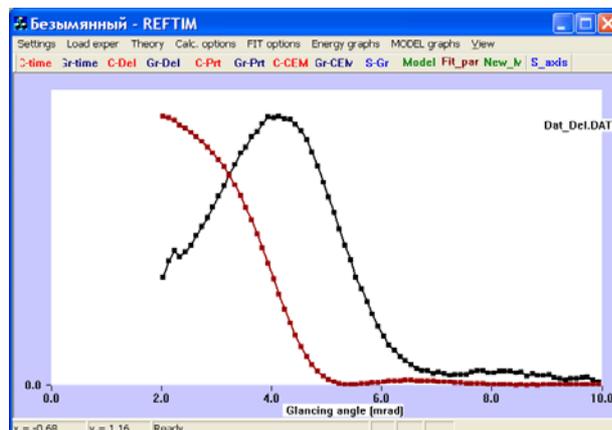
CEMS spectrum should appear on the window if you press **"Gr-CEM".** The experimental data file is just count numbers in column like "**CEMS.dat**" in the "**example**" directory. The calibration parameters are presented in "**FilCEM.INP**" file which you can open and correct by the buttons "**Calc. options**" →"**CEMS options**". After calculation with the chosen model file (like "**m.inf**" in the attached "**example**" directory) by **"C-CEM"** hot-key the theoretical spectra and the multiplet contributions are appeared, the partial spectra are shifted down for clarity.
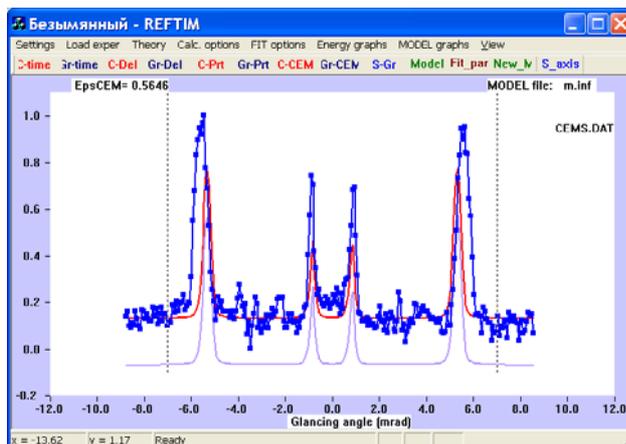
Next you should create a theoretical model for calculations. The sheets for this purpose appear by sequence in **"Settings"** → **"Start from zero"** or by the buttons "**Theory**" → "**Create MODEL**" and suggest a dialog (the model parameters are described in the next paragraph). If you have already prepared the model file you can simply point out the name of the model file in the followed dialog or by buttons "**Theory**" → "**Model file**". In this dialog you also should point out the amount and values of the angles for the reflectivity time spectrum calculations (the angles supposed to be corresponded to the measured time spectra – if you would like to compare theoretical spectra with the experimental ones).

As the example of the model file we give "**m.inf**" in the attached "**example**" directory. You can look at the prepared model file and insert corrections by "**Theory**" → "**Corrections** " or by the hot-key "**MODEL**".

From main menu the buttons "**Energy graphs**" → "**Absorption**" or → "**Reflectivity**" and "**Model graphs**" → "**HFI graph**" or → "**El density**" opens the dialog graph windows which show the energy dependencies of the nuclear resonant spectra of absorption (at grazing angle) or reflectivity and the used depth distribution of HFI or electronic densities. Notice that it works only if you have done at least one calculation of the time spectra or delayed reflectivity curves with your model.

The calculations can be done with the help of the hot-keys buttons: **"C-time"**, **"C-Del"**, **"C-Prt", "C-CEM"** and after that the graphs with the results appeared. Calculations can be performed if the program knows the model file and the needed *.exe files are presented in the used directory.

Buttons: **"Gr-time"**, **"Gr-Del"**, **"Gr-Prt", "Gr-CEM"** – display the graphs of the resultant theoretical spectra or the theoretical spectra imposed on the experimental ones – if you have pointed out the experimental data files at the beginning.

If you are going to investigate just the behavior of the theoretical spectra, then it is not necessary to have experimental data file (but you should create the model file). For this option the number of experimental spectra in consideration should be pointed out as 0.

The following additional options are activated in the program.

When you are in the main window, **the left mouse button** click gives you the x- and y-coordinates of the point (in the left-bottom box).

The right mouse button click activates the dialog window for the correction of the x-, y-scales and for the conversion of the spectra to the logarithm scale and back. The same option with **the right button** is valid for the "**HFI graph**", "**El. density",** "**Energy spectra**" → "**Absorption**" or → "**Reflectivity**"graphs. It is recommended to press "**S_axis**" button after each axis correction in order to save it for the consequent drawing.

Calculation options

You can change the parameters of the computations of the time spectra in the "**TIM09F.INP**" file (It opens by "**Calc. options**" → "**TIM spectra**"). You can choose the type of computation algorithm **Jway**=1 or 2 (matrix or Parratt generalized to the anisotropic case), **ITEST**= 1 or 2 for one step check or for the fit in order to choose the best value of the parameter in consideration (the list of the varied parameters will be described below). You can choose the limits (from **XEXP1** to **XEXP2**) where the numerical comparison of the experimental and theoretical data on the time scale will be done and the "MERIT function" will be calculated. The computation parameter **JFIT** allows to choose the type of the MERIT functions (they are described below). We can fit several time spectra simultaneously, in such case the MERIT functions are summed up with given weights for each time spectrum in consideration, the weights are defined in "**TIM09F.INP**" file, they give a possibility to get the better improvement of the selected spectra. Ab initio the weights are 1 for each spectrum. The computation parameter **EPFIN** points out the limit of the MERIT function improvement for each model parameter variation after which we go to the next model parameter variation. **ISTOP** determines the maximum number of steps for each model parameter variation. **AF** gives the initial constant background in counts which later will be fitted, if **AF**=0 we do not take into account and do not fit the background. The correlation of the model parameter variations can be included: **ICORR1** point out the number of column **ip1** in which all model parameters will be changed simultaneously, if **ICORR1** is negative it points out the number of line **in1** in the list of varied model parameters. **IMAN1** (**IMAN1** = 1,2,3 or 4) determines the way of correlation for this column: **IMAN1=1** means that all column (line) model parameters are equal, **IMAN1=2** means the equal steps, **IMAN1=3** means that the steps are proportional to the value of the model parameter, **IMAN1=4** means that the value of the model parameter is proportional to the old value. If **ICORR1**=0 and **IMAN1**=0 you have no correlations. It is possible up to 5 correlation terms for different columns (lines) simultaneously. The HFI nuclear density distribution can be smoothed over several adjacent sublayers (regularization condition) along the Gauss profile, in this case the calculation parameter **SIG** determines the width (A) of the Gauss regularization, **KP** is number of steps in smoothing.

In the last version REFTIM 7.4 we have inserted the field distribution functions (In previous versions just the correlated with the splitting line broadening was used). It can be the Lorentz or Gauss shape, or determined by the external function from the prepared data file "**DISTR.inf**". The choice is by the parameter **IDIST**=1,2,3 correspondingly in the "**TIM09F.INP**" input file. The number of points in the field distribution description is 2***JAV1**+1, **SD** is the step of the normalized argument, so that for the field deviation ±DBHF(K) from the center of the distribution BHF(K) this argument is equal ±1.

The parameters of the computations of the prompt and delayed curves can be corrected in "**DEL09F.INP**" file (It opens by"**Calc. options**" → "**Del and prompt**"). The calculation parameter **Intexp** = 0 means that there is no experimental data, **Intexp** = 1 means that the

comparison with the experimental curves is supposed. The name of the experimental data file should be on the next line. The calculation parameters now are pointed separately for the prompt and delayed curves, because the calculation time for the delayed curve is much longer than that for the prompt curve. **UT0p**, **Ustepp, Nstepp** and **UT0**, **Ustep, Nstep** are the initial angle, step and the number of points for the prompt and delayed curve calculations respectively. **JFITP** and **JFITD** are the types of the MERIT functions (see below) for comparison of the theoretical and experimental curves (prompt and delayed respectively). **ACOMP1**, **ACOMP2** and **ACOMP1D**, **ACOMP2D** are the limits of the angular range where the comparison of the theoretical and experimental curves is evaluated numerically (for prompt and delayed curves respectively). **EPFINP** and **EPFIND** are the limits for iterations of the MERIT function. **FJEPP** and **FJEPD** are the weights of the **MERIT** functions for the comparison of the theoretical and the experimental curves for the prompt and delayed signal. You can fit just the delayed reflectivity if you put for example **FJEPP**=0 or just the prompt reflectivity if you put for example **FJEPD**=0 (the last one is better to do by just **"C-Prt"** hot-key). For the description of the fall of the reflectivity curve at zero grazing angle divergence of the beam (**DIV**, mrad) and the angle (**ANGBEAM**, mrad), at which the surface completely overheads the beam. (It depends on the surface size.) The parameter **ECYCLE** means the time interval between SR pulses (It is needed if you add to the reflectivity signal the tail from the previous pulse). **TIMINI** and **TIMFIN** the limits (ns) of the experimental time window for the collection of the delayed reflectivity signal.

The service file "**FilCEM.INP**" gives the calculation options for the CEMS spectrum and for calibration of the experimental one (if it is presented). It is supposed that this spectrum is measured (and calculated) in normal geometry from the same sample from which the grazing incidence reflectivity has been measured. The first integer number **NSPCE** number can be 1 or 0 depending if you have the experimental data or not. Then **GS** is a half-width of the source line (for convolution), **EMAX** is a maximum energy half-interval in calculations, **IMM** - number of points for calculation of the CEMS spectrum. After a blank line the name of the file with the experimental data file should be presented. In the next lines the information needed for calibration of the experimental data should be presented: **JEXPC** is the number of experimental points (max 1024), **Hmms** is the field in α-Fe in mm/s, **Hchan** is the field in α-Fe in channels and **chan0** is the number of channel for zero velocity. The numbers **E1lim** and **E2lim** are the limits in the velocity scale (in mm/s) where the comparison of the experimental and theoretical spectra will be evaluated numerically

MODEL PARAMETERS

Here we describe the model parameters that the program has as the input for calculations of the time spectra of reflectivity, of the delayed and prompt reflectivity curves. These model parameters should be presented in the model file "***.inf**" in a definite format. Such model file can be created through the dialog, which is opened by the buttons "**Theory**" → "**Create MODEL**". The name of the prepared file can be also pointed out as the model file for calculations by "**Theory**" → "**Model file**".

First of all you should print in the dialog box the name of the model file, which you are creating (in default it is "**m.inf**"). Then you should point out the amount of the layers NNN characterizing the model stucture, the numbers of the starting and final layer which are repeated in the structure N1N and N2N and the number of repetitions NRN. In default they are 1, 1, 1, 1, i.e. we consider just semi-infinite medium (substrate), no repetitions of layers. The maximum amount of layers in consideration NNN (without repetition) is N24=6**0**. After sub-division of the interfaces into several steps this maximum number could be **300**. The

number of repetitions NRN of chosen sequence of sublayers from N1N up to N2N is arbitrary.

In the same dialog window you should print the number of different types of hyperfine fields (multiplets), which you will use for the calculation of the theoretical spectra of reflectivity NM (the maximum number of NM is N6=8 in this version). In default it is 1. If you have more than 1 multiplet you should print the expected ratio of the partial resonant spectra weights (areas) S(K), K=1,..NM. In default S(1)=1.00, and others are 0. They can be taken from the fit of the measured CEMS spectrum (if you have it).
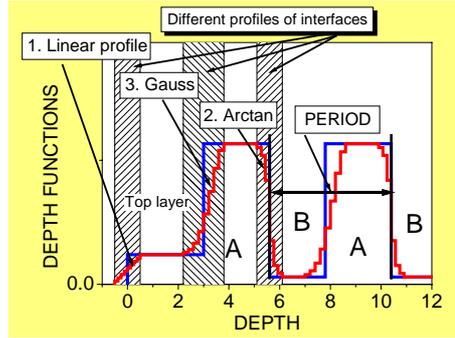
Than you should determine if the hyperfine field orientation will be attributed to the definite layer (IBHF=1) or to the definite multiplet (IBHF=2).

Different types of normalization of the total nuclear density V in the structure is supposed during the fit of the depth distribution of resonant nuclei. It is the parameter INORM. If INORM=1 then V is normalized on the value of the parameter HKMAX (which is 1.00 for pure α-iron with 95% enrichment). That means that the total amount of the resonant nuclei in a layer with maximum concentration of the resonant nuclei is fixed. For INORM=2 V is normalized only when the total density in some layer is larger than HKMAX. If INORM=3 then no normalization is applied. If INORM=4 then the normalization on HKMAX is applied only to the layer where the total density is larger than HKMAX. The parameter ISQ also determine the way of the nuclear density normalization: we can keep the relative amount of nuclei with each kind of hyperfine interaction S(K) (e.g. according to the fit of the CEM spectrum) then you put ISQ=0. If this relative amount is varied freely during the fit of the depth profiles of the nuclear density with definite HFI then we should put ISQ=1, in this case the resultant S(K) are changed during the fit. We can also keep the relative amount of resonant nuclei in each layer, then ISQ=2.
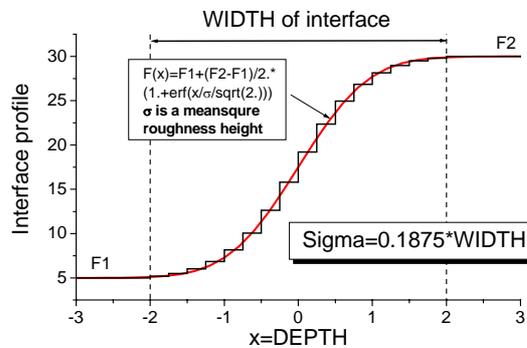
When you press "OK" you get the next dialog sheets in which you should characterize each multiplet used at the beginning (separate sheet for each multiplet).

Each multiplet is characterized by the value of magnetic hyperfine field ($B_{hf}(K)$), width of B_{hf} distribution ($dB_{hf}(K)$), isomer shift (IS(K)), electric field gradient (EFG(K)), additional line width (GRd(K)). Then you point out the angular parameters of B_{hf} orientation which are essential only if you have already choose IBHF=2. They are the polar BE(K) and azimuth GA(K) angles for B_{hf} orientation relative to the surface normal and to the normal to the scattering plane (i.e. BE=90 and GA=90 means that the magnetic hyperfine field is oriented along the beam), relative amount of nuclei with the random in space orientation or random in the surface plane orientation of B_{hf} (parameters RAND(K) and PLAN(K), the sum of them can not be greater than 1). The last box in the dialog sheet is the angular distribution of B_{hf} orientations (parameter WID(K)). In this version of the program just the symmetrical electric field gradient is considered, the main axis of this tensor being coincided with direction of B_{hf}. If IBHF=1 the same angular parameters refer to the definite sublayer and should be pointed out in the description of each sublayer (in the model file these parameters for the case IBHF=1 will be placed in the lines characterising HFI in each sublayer).

Then the separate dialog sheets appear consequently for each layer which you should fill. Each layer is characterized by the thickness H1A(N) (in Angstroms, for the substrate it is unessential) and by the electronic density and photoabsorption (i.e. by the real and imaginary parts of the susceptibility in each layer EL.DENS (in 10^{-6} dimensionless units). The interface profiles are used for the description of the roughness and HFI distributions. The interfaces are characterized by the width of the upper interface - WIDTH - for each layer, by the number of steps KSTEP for the interface function approximation in the reflectivity calculations. The type of function KIND1, KIND2 is used for the real and imaginary parts of the electronic susceptibility respectively for each interface

Different profiles of interfaces

1. Linear profile

3. Gauss

2. Arctan

PERIOD

DEPTH FUNCTIONS

Top layer

0.0

A   B   A   B

0   2   4   6   8   10   12

DEPTH

The value of these parameters KIND1, KIND2 means: 1 – linear function, 2 – arctangent, 3 – half Gauss profile, 4 –Gauss profile, 5 – error function. The characteristic dimensions of the interface functions 2, 3 and 4 (e.g. the width of Gauss function or the steepness of arctangent) are given by ZD1, ZD2 for the real and imaginary parts of the susceptibility. The steepness of the error function is fixed.

WIDTH of interface

F2

30

Interface profile

$F(x)=F1+(F2-F1)/2.*$
$(1.+erf(x/\sigma/sqrt(2.)))$
**σ is a meansqure
roughness height**

25

20

15

10

Sigma=0.1875*WIDTH

5

F1

-3   -2   -1   0   1   2   3

x=DEPTH

In this case the interface WIDTH is connected to the meansquare roughness height σ, which is often used in the reflectivity calculation as the suppression factor $e^{-2k^2\sigma^2}$ at each boundary by the relation σ= 0.1875*WIDTH. (the factor was adjusted empirically). Nuclei with each type of HFI can be distributed by their own manner in the multilayer structure. The depth distribution function is determined by the matrix V[N,K] which is actually the density of nuclei (normalized to the density of Fe atoms in pure α-Fe) with the definite type K of HFI in the layer with number N. In the dialog "**Theory**" → "**Create MODEL**" we fill the values of V[N,K] for each layer consequently: N is fixed, K=1, .. NM, accordingly to the appearing sheets. For the description of the HFI distributions in the interfaces the same width and number of steps is used as it has been chosen for the electronic susceptibility description (parameters WIDTH and KSTEP). However the kind of function in each interface can be chosen independently. That is a matrix of integer numbers KINDV(N,K) (K=1,…N6, N=1,…NNN) for each type of HFI and each interface. The meaning of these numbers 1, 2, 3, 4 and 5 is the same as for the description of the susceptibility interface functions. Accordingly the matrix ZDV(N,K), (K=1,…N6, N=1,…NNN) should be filled, the elements of which describe the parameter of the interface functions (for 2,3,4 types of functions) for each kind of multiplet in each layer. The same sequence is valid for the filling the matrix element of KINDV(N,K) and ZDV(N,K). In the same layer sheets you put the values of the angular parameters for each layer (BE, GA, RAND, PLAN, WID) if in the first sheet you have chosen the special parameter IBHF=1.

The dialog for the model creation is finished by the suggestion to save the prepared file. You should tick off the box in order to continue the calculation of the reflectivity with the created model file.

You can look at the prepared model file and insert corrections by "**Theory**" → "**Corrections** " or by the hot-key "**MODEL**". The dialog graph windows "**Energy spectra**" → "**Absorption**" or → "**Reflectivity**" and "**Model graphs**" → "**HFI graph**" or → "**El density"** show the energy dependencies of the nuclear resonant spectra of absorption (at grazing angle) or reflectivity and the used depth distribution of HFI or electronic densities. Notice that it works only if you have done at least one calculation of the time spectra or delayed reflectivity curves with your model.

As the example of the model file we give "**Exercise.inf**".

```
   LAYSPC.INF
              PARAMETERS OF SUBLAYERS
   3    NNN - number of sublayers (max N24=60, including substrate
   1  2    N1N,N2N - limits of repeated layers (from - to, including)
   5       NRN - number of repetitions
   3   INORM=1 - V is norm,=3 -is not ; =2- V norm.from top,=4 for  one larger layer
   1  ISQ =0- V is recalculated,=1- S((K) are recalcul.,=2 -keep   HK
  1.00000    HKMAX - max value of nuclear density (1.for pure a-57Fe)
   2    NM - number of multiplets (max N6=8)
   2       IBHF=1: Bhf-EFG we read from here; IBHF=2: BE-WID refer to each multiplet
      H1A       EL.DENS.    ZD1     ZD2    WIDTHKIND1 KIND2 KSTEP
 1  30.0000  -10.000  0.200   0.000    0.000   30.000   1    1    0
 2  85.0000  -14.000  0.700   0.000    0.000   30.000   5    5    0
 3 100.0000   -5.000  0.020   0.000    0.000   60.000   5    5    6
              V[N,K]   MULTIPLET DENSITIS and if IBHF=1: BE   GA  Rand Plane  WID
 1   0.05000 0.35000 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000   0.0   0.0 0.000 0.000   0.0
 2   0.70000 0.10000 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000   0.0   0.0 0.000 0.000   0.0
 3   0.00000 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000   0.0   0.0 0.000 0.000   0.0
          Corrected S(K)-ratio of partial spectra areas
 4   0.76250 0.23750 0.00000 0.00000 0.00000 0.00000 0.00000 0.00000
          Hyperfine parameters for each multiplet
 5   33.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000   Bhf(T)
 6    2.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000   dBhf(T)
 7    0.0000  0.3000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000   IS(mm/s)
 8    0.0000  0.6000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000   EFG(mm/s)
 9    0.0000  0.1000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000   GRd(mm/s)
       Angular parameters for each multiplet if IBHF=2
11    0.0    0.0    0.0    0.0    0.0    0.0    0.0    0.0  BE
12    0.0    0.0    0.0    0.0    0.0    0.0    0.0    0.0  GA
13   0.000  0.000  0.000  0.000  0.000  0.000  0.000  0.000  RAND
14   0.000  0.000  0.000  0.000  0.000  0.000  0.000  0.000  PLAN
15    0.0    0.0    0.0    0.0    0.0    0.0    0.0    0.0  WID
 Kind of interface function for each multiplet in each layer KINDV[N,K],N=1,NNN,K=1,N6
 1    1    1    1    1    1    1    1    1
 2    1    1    1    1    1    1    1    1
 3    1    1    1    1    1    1    1    1
 Parameter of interface function for each multiplet in each layer ZDV[N,K]
 1   0.000  0.000  0.000  0.000  0.000  0.000  0.000  0.000
 2   0.000  0.000  0.000  0.000  0.000  0.000  0.000  0.000
 3   0.000  0.000  0.000  0.000  0.000  0.000  0.000  0.000
```

It represents two layers, the first one having the thickness 30 A and electronic susceptibility χ^{el}=(-10.0+0.2 i) *$10^{-6}$and the other layer (85 A) having susceptibility χ^{el}=(-14.0+0.7i)*10^{-6} , these two layers are repeated 5 times, on the nonresonant substrate (χ^{el}=(-5.0+0.02 i)*10^{-6}). The layers have no interfaces, but the substrate surface has. See the picture of the depth profile of the electronic susceptibility below. You get it after calculation of the any curves by "**Model graphs**" → "**El density"**. There are two kinds of hyperfine interaction (HFI): NM=2, the first one is characterize by B_{hf}=33T with the distribution width 2.T, the other one is characterized by 0.6 mm/s quadrupole splitting, 0.3 mm/s isomeric shift and 0.1 mm/s line broadening. The nuclear resonant isotope with the first type of HFI is presenting mainly in the layer number 2 with 0.7 factor of the density of the ^{57}Fe nuclei in 95 % enriched pure α-iron (V[2,1]=0.7), the first layer contains 0.05 portion of HFI of the first kind and 0.35 portion of HFI of the second kind (V[1,1]=0.05, V[1,2]=0.35). Mention that the

parameter INORM is chosen as 3, which means that no additional normalization of the absolute values of the elements of V[N,K] will be done. Substrate is nonresonant. The angular parameters for the hyperfine field is attributed to the multiplets (not to the layers) IBHF=2, and they are characterized by the definite azimuth and polar angles Beta=0°, Gamma=0°, which means that B_{hf} is oriented perpendicular to the surface plane as well as the main axis of the electric field gradient. The picture of the resonant spectrum splitting (theoretical CEM spectrum) and depth profiles of nuclear density with different HFI is presented below for this model (they are vertically shifted for clearity). You get it after calculation of the any curves by "**Model graphs**" → "**HFI graph**"
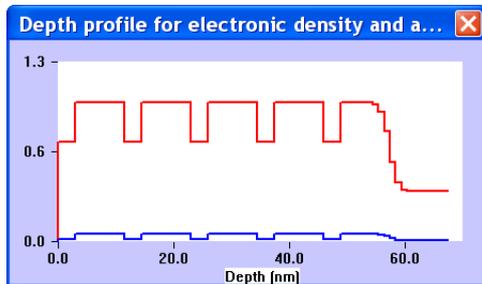

Red line is the normalized depth profile of -Re χ (z),
blue line is the depth profile of Im χ (z).

Theoretical CEM spectrum at normal geometry
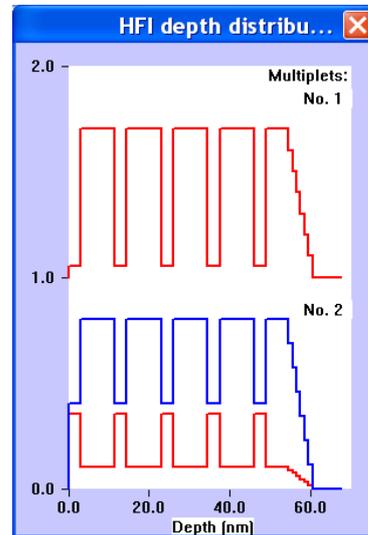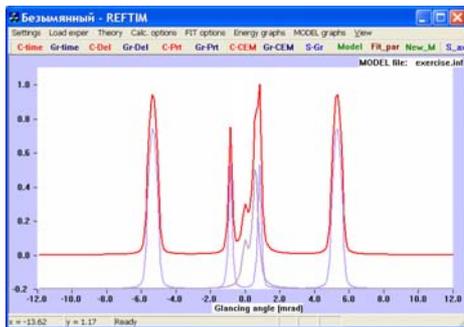
Red lines are the depth distribution of the ^{57}Fe isotope density characterized by the HFI No.1 (shifted up) and No.2; blue line represents the depth profile of the total nuclear isotope density.

The corresponding time spectra at the Bragg maxima and delayed (blue) and prompt (red) curves (in logarithm scale).
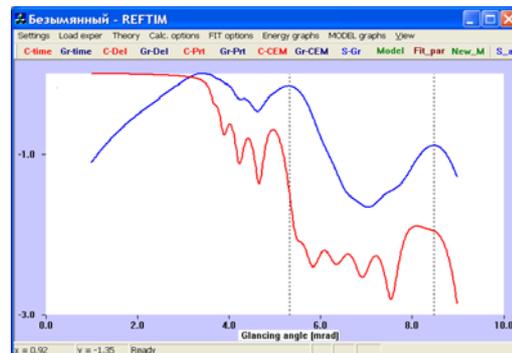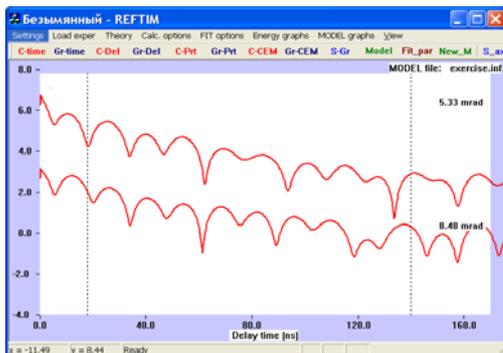
If you would like to change the model essentially (e.g. to change the number of the layers NNN or the number of multiplets NM for the description of the spectra), it is recommended to go again to the dialog "**Theory**" → "**Create MODEL**", and from the begining change the numbers NNN and NM. In such a way you will be sure that the new model file will have the right format for calculations. By handle change of the model file you should be careful about the number of lines in the description of the sublayer thicknesses, HFI

depth distribution description, kind of interface function for each multiplet in each layer and parameters of interface function for each multiplet in each layer.

Some limitations (just due to the memory restriction during computation):

The maximum amount of the time spectra (calculated for different angles) for simultaneous consideration is **4**.

The maximum amount of different HFI for ^{57}Fe nuclei in consideration is **8**.

The maximum amount of sublayers in consideration (without repetition) is **60**. After sub-division of the interfaces into several steps this maximum number could be **300**. The number of repetitions of chosen sequence of sublayers is arbitrary.

MERIT FUNCTIONS

For the dependencies which you prefer to watch in logarithmic scale the choice of the MERIT function is not simple. In order to enhance the contribution of the "tails" of the curves to the mean-square deviation of the theoretical dependence from the experimental one it was suggested to use the different power **n** of the denominator of the MERIT function (We designate it as **EPS**):

$$\text{EPS} = \sqrt{\sum_i \frac{(y_i - t_i)^2}{y_i^n}} \Big/ \sqrt{\sum_i \frac{1}{y_i^n}} \tag{1}$$

where n=1,2,3 … and respectively **JFIT**=1,2,3, ….. (or **JFITP and JFITD**) for the choice of the MERIT functions in "**TIM09F.INP**" or "**DEL09F.INP**". In (1) y_i and t_i are the values of the normalized experimental and theoretical points on the graphs, N is the number of points in comparison.

In the paper [A.Segmuller, Int.conf. "Modulated structures", New-York,1979. p.78-80] an another criteria were used. According to this paper we suggest also to use the following MERIT functions.

$$\text{EPS} = \frac{1}{\sqrt{N}} \sqrt{\sum \frac{(y_i - t_i)^2}{(y_i + t_i)^2} + \sqrt{\frac{\sum(y_i - t_i)^2}{\sum y_i^2}}}, \quad \text{JREP}=11 \tag{2}$$

$$\text{EPS} = \sqrt{\frac{\sum(y_i - t_i)^2}{\sum y_i^2}}, \quad \text{JREP}=12 \tag{3}$$

$$\text{EPS} = \frac{1}{\sqrt{N}} \sqrt{\sum \frac{(y_i - t_i)^2}{(y_i + t_i)^2}}, \quad \text{JREP}=13. \tag{4}$$

JREP=13 adjusts preferably the "tails" of the curves, JREP=12 adjust better the largest part of the curves. The most suitable results in many cases we got with the function, corresponding to JREP=11.

The choice of the criteria depends on the appearance of the curve. We can choose the MERIT function by comparing the influence of the different MERIT functions on the quality of the obtained result.

MODEL PARAMETER VARIATIONS

The calculated spectra depend on the enormous amount of the structure and HFI parameters. If you want to follow the influence of each parameter on the spectra you can just open the model file by "**Theory**" → "**Corrections** " or by the hot-key "**MODEL**", change the selected parameter and make calculations by **"C-time"**, **"C-Del", "C-Prt"** or **"C-CEM"**. You can see in the graph simultaneously the initial (first one for the session) theoretical spectra (thin lines) and the new one (thick lines) : solid red lines are for time spectra and prompt reflectivity, or blue curves are for delayed reflectivity.

The same result will be if you choose ITEST=1 (one step check start) in the "**TIM09F.inp**" file by "**Calc. options**" → "**TIM spectra**" → "**File**" or "**Dialog**", but in this case you should previously point out the chosen parameter and step in the file "**Next.inp**" which is opened e.g. by the hot-key "**Fit_par**". The way how you point out the selected parameter and step (or sequence of parameters and steps) in this file is the same as during the fit procedure which we describe below, but with ITEST=1 you will have just one step check.

By default ITEST=2. (You also can choose ITEST=2 in "**TIM09F.inp**" by "**Calc. options**" → "**TIM spectra**" → "**File**" or "**Dialog**"). That means that the iteration procedure for the improvement of the agreement between theoretical and experimental spectra for each chosen parameter will be performed. We use the consequent one-parameter descending. The sequence of the fitted parameters is determined by the file "**Next.inp**". It is opened by the hot-key "**Fit_par**". From the beginning the "**Next.inp**" file contains just two lines, the first one is 1 and the second line is six numbers 0. (That line should end any sequence of parameters.) This initial "**Next.inp**" means that there is no any parameter for the fit. For the fit we should include the additional lines in "**Next.inp**": one additional line for each parameter which we want to vary.

In the last version of the program the simplest way for the filling the "**Next.inp**" file is suggested. You should press "**FIT options**" → "**List_par**" and all parameters, which you are able to vary in your structure and HFI model, will be listed in the opened file. Each parameter is characterized by the two numbers, which are written at the beginning of each line. They are the number of layer (the numbering is from the surface to substrate) or the number of HFI parameter and the type of the parameter (e.g. second number 1 means the thickness of the corresponding layer, 2 means the real part of susceptibility etc.) or the number of HFI. The third column in the file "**List_par.res"** gives the value of all these parameters in your model. You just print the nonzero initial step (the 4th number in each line) for each parameter, which you want to vary during the fit and when you close "**List_par.res"** (with saving the corrections) the "**Next.inp**" file will be created automatically.

The last 3 numbers in each line of "**List_par.res"** file (and "**Next.inp**" file) give the possibility to connect the variation of the chosen parameter with the variation of the additional parameter. By default they are 0 0 1.0. That means that there is no correlated variation. Two integer numbers designate the connected parameter. The last number is the factor multiplying the step of the first parameter variation for the obtaining the step for the variation of the connected parameter. If this number will be –1.0 the variation of the connected parameter will be opposite to the variation of the first parameter. It is important e.g. when you would like to change the thickness of one layer in the period, but you want to keep the value of the period.

It is recommended to look at the obtained "**Next.inp**" file (by "**FIT options**" → "**Check list**" or by the hot-key "**Fit_par**") for some corrections. For example you can change the sequence of lines in order to change the sequence of the parameter variations or to repeat some lines or group of lines for the repetition of the previous fit etc. Remember that the last line in the "**Next.inp**" file should be six 0 numbers.

On the way "**FIT options**" → "**Fitting**" you have different options: "**Time spectra**", "**Prompt curve**", "**Delayed and prompt curves**", "**Joint_FIT**". That means that you can choose to fit just the time spectra (it could be done with different weights for time spectra measured for different grazing angles determined in "**TIM09F.inp**" and over different time range), just the prompt reflectivity curve, both prompt and delayed reflectivity curves (if you point out JFEPP=0 in "**DEL09F.inp**" – see section 2 it will be the fit just of the delayed curve) and at last the more effective option "**Joint_FIT**" is for simultaneous fit of all kind of measured data: both reflectivities and time spectra. If you do the fit from this way "**FIT**" → "**Fitting**" you will be able to watch the main window during the fit and follow the obtaining variations of the curves by pressing **"Gr-time","Gr-Del"** or **"Gr-Prt"** from time to time. You also should not forget to do that at the end of calculation !. When the fit has been finished the old and new value of the MERIT function appears in the upper-left corner of the screen.

The fit of any curve can be initiated simply by the hot-key **"C-time", "C-Del", "C-Prt"** or **"C-CEM"** and mark the box in the dialog window. This dialog window appears only when the "**Next.inp**" file contains the lines with selected parameters for fit. Notice that when you use **"C-time", "C-Del", "C-Prt"** or **"C-CEM"** for the fit option it is not possible to follow the consequent transformation of the theoretical curves during the optimization of the selected parameter value on the graphs by press the button **"Gr-time","Gr-Del", "Gr-Prt"** or **"Gr-CEM"** respectively. However the final result appears on the screen automatically. For the joint fit just the way "**Fitting**" → "**Joint_FIT**" works.

In spite that we use just one parameter descending there are a lot of other options for correlation of the parameter variations which expand the possibilities of the program REFTIM. They are:

♦ the correlation between parameters in each layer or for each type of parameter,

♦ the normalization of the HFI distribution on the maximum density of nuclei in the structure or in each layer,

♦ the normalization of each HFI relative weight (when you can compare with e.g. multiplet areas in the absorption spectrum),

♦ the smoothing of the depth distribution functions for each multiplet or for all multiplets,

♦ you can attribute the angles of the HFI orientation for a given layer or to the given multiplet.

RESULTS

After each calculation or fit procedure the new model file "**LAYSPC.RES**" is created corresponding to the best obtained result for the varied parameters. You can look and save it for the memory or for the next calculations by the button "New_M". You can watch also on the graphs of the last obtained electronic density depth distribution and photoabsorption compared with the initial ones by "**Model graphs**" → "**El density"** or the obtained and initial depth distribution of HFI nuclear density for each multiplet by "**Model graphs**" → "**HFI graph**". The energy dependencies of the nuclear resonant spectra of absorption (at grazing angle) or reflectivity is presented by "**Energy spectra**" → "**Absorption**" or → "**Reflectivity**".

After each calculation the resultant spectra are saved in "***.RES**" files. For the time spectra they are "**BUFFTN1.RES**", "**BUFFTN2.RES**", … files. For the energy dependencies of reflectivity they are "**BUFFEN1.RES**", "**BUFFEN2.RES**", … files. For the prompt dependencies it is "**RELAY.RES**" file in absolute values and superimposed on the experimental curve in "**PROMPT2.RES**". For the delayed curves it is "**DELAY2.RES**" file.

"**CEMfit.res**", "**CEMorig.res**" files presents of the results of the CEM spectrum calculation or fit, in the last one the data is normalized on the effect value. The model of HFI distribution for each multiplet is presented in the "**MODELN.RES**" file, the model of the electronic density is saved in the "**EL_MOD.RES**". The absorption resonant spectra for two polarization of the incident radiation are in the "**E_spcN.RES**". If you made the angle variation the final angle is put to "**ANGLE.RES**", the old one is kept in "**OLD_ANG.RES**". The control parameters of computations are placed in "**TIM09F.res**", "**DEL09F.res**", "**REF09F.res**" and "**CEM09.res**" for each computation codes "**TIM09F.exe**", "**DEL09F.exe**", "**REF09F.exe**" and "**CEM09F.exe**" correspondingly. For the check of the procedure the non-normalized and non-averaged time spectra for first angle of calculation and the first delayed curve is saved in "**ttestF.res**" and "**timtesF.res**" for "**TIM09F.exe**" and "**DEL09F.exe**" respectively.

THE SIMPLEST EXAMPLE

We have prepared the "experimental" data by calculation of the prompt and delayed angular curves of the reflectivity ("**Dat_Del.dat**"), CEMS spectra ("**CEMS.dat**") and of the time spectra for the angle 2.2 mrad ("**Dat_Tim.dat**") for the simplest model ("**m_dat.inf**"). Then we slightly change the value of the hyperfine field and width of its distribution and save the corrected model in the file "**m.inf**". The whole set of the needed for the exercise files is placed to a subdirectory "**example**".

Being in this directory we press "**REFTIM.exe**", "**Settings**" → "**Get previous settings**" and get the "experimental" time spectrum on the screen. We can check that we are going to make calculations with the model file "**m.inf**" and for one grazing angle 2.2 mrad by "**Theory**" → "**Model file**", the model file "**m.inf**" can be watched by the hot-key "**MODEL**". The delayed and prompt "experimental" curves appear on the screen by **"Gr-Del"** or **"Gr-Prt"** button. **"Gr-CEM"** button presents the "experimental" CEM spectrum. The calculations by **"C-time"**, **"C-Del"**, **"C-CEM"** or **"C-Prt"** show that the used model is far from the "experimental" case.

For the fit procedure we press "**FIT options**" → "**List_par**" and in the opened file "**List_par.res**" we put the initial step (e.g. 1.) for the value of the hyperfine field and width of its distribution. When we close (with saving) the file "**List_par.res**" and the new file "**NEXT.INP**" is created. You check it by the hot-key "**Fit_par**":
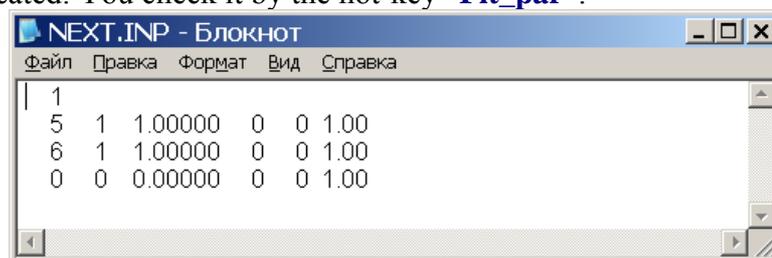
It means that at first the parameter 5 1 (the value of B_{hf}) will be varied during calculation for the search of the optimum value and after that the parameter 6 1 (the width of the hyperfine field distribution) will be varied. By default the type of this distribution is Gauss function according to IDIST=1, this parameter can be changed in the "**TIM09F.INP**" file by buttons "**Calc. options**" → "**TIM spectra**". Then we can start "**C-Time**", agree to fit by the mark of the box in the dialog window and see the improvement of the theoretical spectrum. The new model we should save by the button "New_M" with the new name, say "**m1.inf**". You can also check (by "**C-CEM**") that the CEM spectrum shows the better agreement with the experimental data and by the button "**C-Del**" the improvement of the delayed curve. Then procedure "**C-Time**" can be repeated for the better result.