

DeConvEELS

(Software Monochromator)

DigitalMicrograph Plugin

for

Electron Energy Loss Spectrum Deconvolution

User's Guide

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1. Introduction

DeConvEELS is the software for rectifying Electron Energy Loss Spectrum (EELS) by deconvoluting it with a low-loss or zero-loss spectrum. This is a software monochromator, since it will eliminate energy spread of incident electrons. The DeConvEELS software consists of a suite of scripts and a Plug-In for use in Gatan's DigitalMicrograph.

This DeConvEELS User's Guide is written to provide information on the basic functions of the DeConvEELS software, a procedure for installation of the Plug-In, some general tips on operation and troubleshooting. This Guide assumes the user is familiar with image manipulation within DigitalMicrograph as well as Windows operating system.

1.1 Features

1.1.1 Advanced deconvolution algorithms

The DeConvEELS software uses a Maximum Entropy Method (MEM) or the Richardson-Lucy Algorithm (RLA) for a spectrum deconvolution. Both procedures will work on a usual spectrum data as well as two-dimensional spectrum data obtained by using a CCD camera. The DeConvEELS software gives an ideal result for a good signal-to-noise data within a reasonably short processing time.

1.1.2 Easy-to-Use User Interface

The DeConvEELS software is easy to use and normally works with default setups. However, the user can change its setups easily within a custom-made setup dialog (See **sections 2.2 and 2.3**).

NOTE An image (spectrum) of *any size* can be handled (not necessary to be a power of two).

However, a low-loss spectrum and a spectrum to be processed should have the same size.

TIPS Create a new image of power of two from a selection. You can easily create a new image of power of two using a ROI selection as follows:

1. Select an area of power of two using a ROI tool.
2. Use a regular Copy short cut: Ctrl + C.
3. Use a modified Paste short cut: Ctrl + Alt + V.

1.2 Deconvolution Overview

1.2.1 Observed Spectrum and Deconvolution

An observed spectrum is a sum of ideal spectrums, each of which corresponds to specific electron energy before inelastic interaction. Therefore, an observed spectrum $h(x)$ is given by a convolution of an ideal spectrum $f(x)$ and a low-loss spectrum $g(x)$:

$$h(x) = g * f(x) = \int g(x-t)f(t)dt .$$

This convolution integral is written as a product in the Fourier space as

$$H(\xi) = G(\xi) \cdot F(\xi) .$$

Therefore, if we can ignore a noise, the ideal spectrum is easily restored by taking an inverse Fourier transform of $H(\xi)/G(\xi)$, the observed spectrum's Fourier transform divided by the low-loss spectrum's Fourier transform. However, this Fourier deconvolution does not work for a real spectrum, since we cannot ignore the noise.

1.2.2 Maximum-Entropy Method

Maximum Entropy Method (MEM) tries to find a solution by using a concept of entropy.

$$S(f) = -\sum_i f_i \log(f_i) , \text{ where } \sum f_i = 1, f_i \geq 0$$

Here, the following constrained entropy is minimized

$$Q(f) = \alpha S(f) - L(f)$$

where L(f) is called a Likelihood defined by

$$L(f) = \frac{\chi^2(f)}{2} = \frac{1}{2} \sum_i \frac{(h_i - (g * f)_i)^2}{\sigma_i^2}$$

There are several ways to minimize the constrained entropy. We choose Collins' iterative procedure, where the following Shannon-Jaynes entropy is employed

$$S(f) = -\sum_i f_i \log(f_i/m_i) , \text{ where } \sum m_i = 1, m_i \geq 0 \text{ (measure)}$$

Here, we use a previous solution as the measure. Therefore, we have the following iterative formula

$$Q(f^k) = -\sum_i f_i^k \log(f_i^k / f_i^{k-1}) - \lambda \sum_i \frac{(h_i - (g * f^k)_i)^2}{\sigma_i^2} , \text{ where } f_i^0 = 1$$

1.2.3 Richardson-Lucy Algorithm

Richardson-Lucy Algorithm (RLA) tries to find a solution by the following iterative formula:

$$\psi^{k+1}(\xi) = \psi^k(\xi) \int \frac{P(x, \xi) \phi(x)}{\int P(x, \xi) \psi^k(\xi) d\xi} dx \quad \text{using } \psi^0(\xi) \equiv 1$$

This algorithm is explained as follows by using formulas in statistics. We know an kernel function P(x) and observe a data $\phi(x)$, and the data is given by the following convolution integral with an unknown function $\psi(x)$:

$$\phi(x) = \int P(x - \xi) \psi(\xi) d\xi = \int P(x, \xi) \psi(\xi) d\xi$$

If we can find an inverse-kernel function Q(x), we can estimate the unknown

function $\psi(x)$ by using the given data $\phi(x)$

$$\psi(\xi) = \int Q(\xi - x)\phi(x)dx = \int Q(\xi, x)\phi(x)dx$$

In the Richardson-Lucy algorithm the inverse-kernel function is iteratively estimated by a previous solution by using the Bayes' theorem as follows:

$$Q^k(\xi, x) = \frac{P(x, \xi)\psi^k(\xi)}{\int P(x, \xi)\psi^k(\xi)d\xi}$$

Thus, we have the Richardson-Lucy formula given above.

1.2.4 Convergence Criteria

For the iteration processes, it is very important to use a useful convergence criterion. The following shows some criteria used in the DeConvEELS:

$$\text{R-factor: } R = \sum |\phi_{cal}(i) - \phi_{obs}(i)| / \sum \phi_{obs}(i)$$

$$\text{Variance } (\chi^2): \chi^2 = \sum (\phi_{cal} - \phi_{obs})^2 / N$$

$$dQ: dQ = Q(f^k) - Q(f^{k-1}) \text{ (applicable to MEM only)}$$

The useful convergence criterion may be a difference of R-factor at each cycle, not the R-factor itself. The Likelihood (χ^2) is always decreases at each cycle, while the constrained entropy Q will oscillate at certain point of cycle. Thus, dQ may become negative, when a selected convergence factor is too large. Therefore, when iteration is stopped by dQ criteria, you may have to recalculate using a smaller convergence factor. In this way dQ is another useful criterion for the case of MEM processing.

Note dQ is not a criteria to stop iteration but a measure to check an *inappropriate* convergence speed (convergence factor).

1.2.5 Important Note

Theoretically, a constant background will not affect the result of deconvolution, since a convolution between a constant value and any kernel is a constant. On the other hand, both techniques of deconvolution used in this plug-in always give a positive result all over the region, and guarantee a non-negative result. However, they are apt to give an overshoot at the edges of a sharp peak, or the edge of a steep onset of spectrum. Even though this overshoot will not become negative, but gives a deep valley if there is an appreciable background. Thus, *it is advisable to subtract an unnecessary constant background before performing deconvolution.*

2. DeConvEELS

The DeConvEELS software is a plugin for the Gatan's DigitalMicrograph, and thus only works under the DigitalMicrograph. The DeConvEELS consists of a suite of menu commands and a dynamic link library.

2.1 DeConvEELS Commands

The DeConvEELS commands will appear under the EELS menu as shown below.

Figure 2-1

DeconvEELS commands under the EELS menu



Various control parameters for Maximum Entropy or Richardson-Lucy deconvolution processing will be specified through a dialog that will be opened by selecting a corresponding command. When the command is launched with the ALT-key down, you can bypass these setup dialogs and use previous setup parameters.

2.2 Maximum Entropy Setup

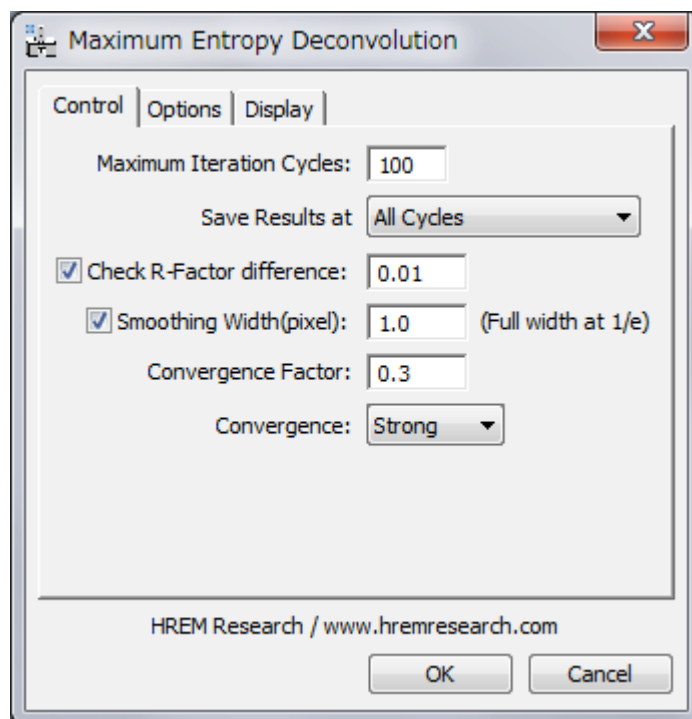
The Maximum Entropy command will open a dialog to control the processing and display the results.

2.2.1 Control tab-panel

The front most tab-panel has some general controls for deconvolution processing.

Figure 2-2

Maximum Entropy Deconvolution (Control Tab)



Maximum Iteration Cycles: Maximum number of iteration cycles.

Save Results at: This controls a frequency of saving the results during the progress of deconvolution. A two-dimensional deconvolution usually requires a plenty of memory to save all the intermediate results. You can select from

All Cycles/Every Two Cycles/Every Four Cycles/Every Eight Cycles /R-Factor (1/3, 1, 3).

Check R-Factor difference: When selected, the iteration will stop when the difference of R-Factor divided by the R-Factor itself becomes smaller than the value specified here.

$$\Delta R = (R_{\text{previous cycle}} - R) / R$$

Smoothing Width (Full width at 1/e): When selected, the raw data will be smoothed out by convoluting a Gaussian with a specified half-width (in pixels). This is especially effective for a noisy spectrum

Convergence Factor: This controls a speed of convergence. The default value is 0.3. A smaller value results in a slower convergence.

Convergence: There is a selection of convergence between Strong and Weak. The default setting is Strong.

NOTE: All the convergence criteria will be used logical “OR,” so iteration will stop when one of the criteria is satisfied.

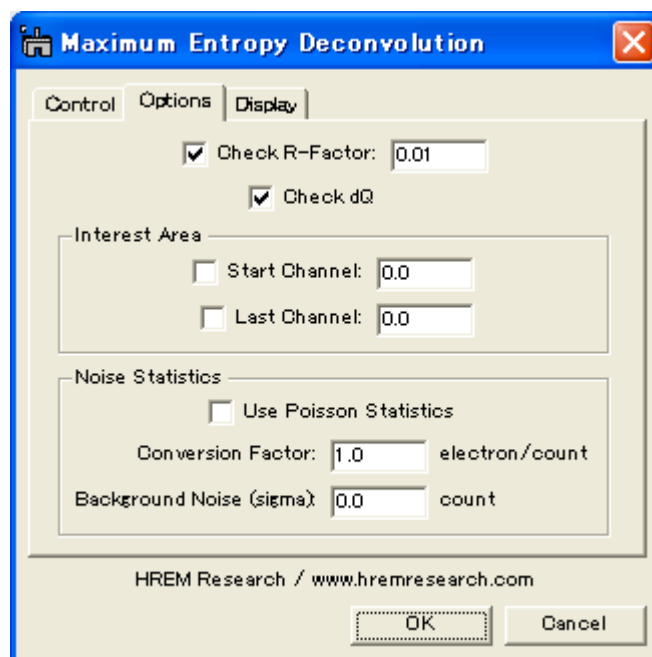
Weak convergence changes the convergence factor automatically, when the difference of Q value starts oscillating. Therefore, dQ criteria in the Options tab has no effect for the **Weak** convergence.

2.2.2 Options tab-panel

The next tab-panel is designed for specifying some optional controls for deconvolution processing. These controls will be useful for deconvolution of a low-loss area, where a zero-loss is far dominant than the loss-loss signal.

Figure 2-3

Maximum Entropy Deconvolution (Options Tab)



Check R-Factor: When selected, the iteration will stop when R-Factor becomes smaller than the value specified here. It is not recommended to activate this criterion, when you don't know an approximate noise level.

Check dQ: When checked, the iteration will stop when the difference of Q value starts oscillating.

Interest Area: You can specify an interest area (ROI; a region of interest) of your spectrum. This is useful for deconvolution of a low-loss area, since convergence criteria will not be affected by the dominant zero-loss area.

Note Start and Last channels of the “Interest Area” will be read from Interest Area “ROI” (see 4.3.2).

Noise Statistics: You can explicitly use Poisson Statistics for noise distribution. In this case you have to specify Conversion Factor in order to convert a signal count to a number of electrons. You can specify also a constant Background Noise (Gaussian) such as a read-out noise.

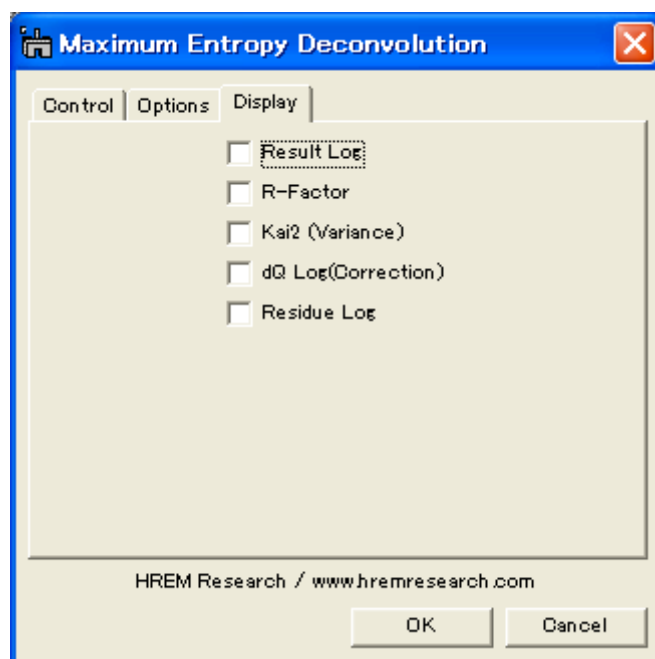
Note When “Use Poisson Statistics” is deselected, a constant noise model is used for the EELS signal. Although this is not a real noise model for EELS signal, it seems to work very well for a core-loss signal.

2.2.3 Display tab-panel

The last tab-panel is designed for selecting the various results to be displayed at the end of processing.

Figure 2-4

Maximum Entropy Deconvolution (Display Tab)



Result Log: All the intermediate results specified by “Save Results at” will be saved.

R-Factor: History of R-factor.

χ^2 (Variance): History of squared sum of differences.

dQ (Correction): History of sum of absolute corrections.

Residue Log: All the intermediate difference between an observed image and an estimated image (convolution between the probe and deconvolution) will be saved at specified cycles.

2.3 Richardson-Lucy Setup

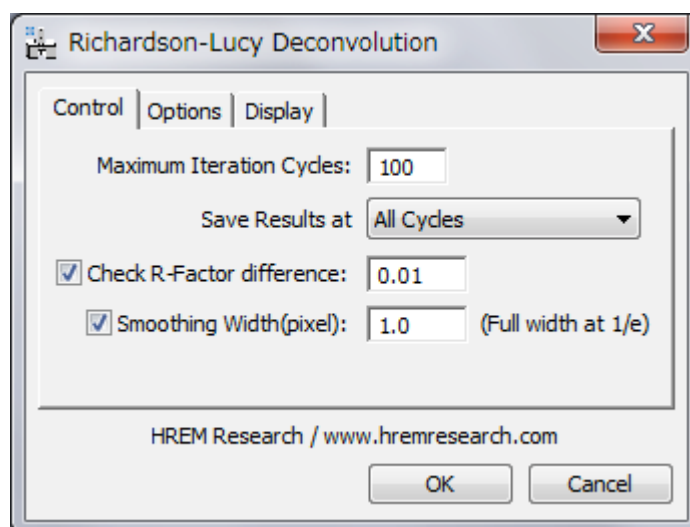
The Richardson-Lucy command will open a dialog to control the processing and display the results.

2.3.1 Control tab-panel

The front most tab-panel has some controls for deconvolution processing.

Figure 2-5

Richardson-Lucy Deconvolution (Control Tab)



Maximum Iteration Cycles: Maximum number of iteration cycles.

Save Results at: This controls a frequency of saving the results during the progress of deconvolution. A two-dimensional deconvolution usually requires a plenty of memory to save all the intermediate results. You can select from

All Cycles/Every Two Cycles/Every Four Cycles/Every Eight Cycles /R-Factor (1/3, 1, 3).

Check R-Factor difference: When selected, the iteration will stop when the difference of R-Factor divided by the R-Factor itself becomes smaller than the value specified here.

$$\Delta R = (R_{\text{previous cycle}} - R) / R$$

Smoothing Width (Full width at 1/e): When selected, the raw data will be smoothed out by convoluting a Gaussian with a specified half-width (in pixels). This is especially effective for a noisy spectrum

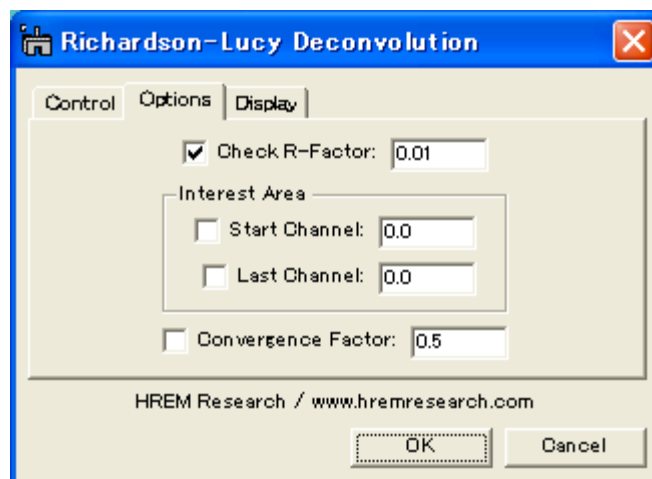
NOTE: All the convergence criteria will be used logical "OR," so iteration will stop when one of the criteria is satisfied.

2.3.2 Options tab-panel

The next tab-panel is designed for specifying some optional controls for deconvolution processing. These controls will be useful for deconvolution of a low-loss area, where a zero-loss is far dominant than the loss-loss signal.

Figure 2-6

Richardson-Lucy Deconvolution (Option Tab)



Check R-Factor: When selected, the iteration will stop when R-Factor becomes smaller than the value specified here. It is not recommended to activate this criterion, when you don't know an approximate noise level.

Interest Area: You can specify an interest area (ROI; a region of interest) of your spectrum. This is useful for deconvolution of a low-loss area, since convergence testing will not be affected by the dominant zero-loss area.

Note Start and Last channels of the "Interest Area" will be read from Interest Area "ROI" (see 4.3.2).

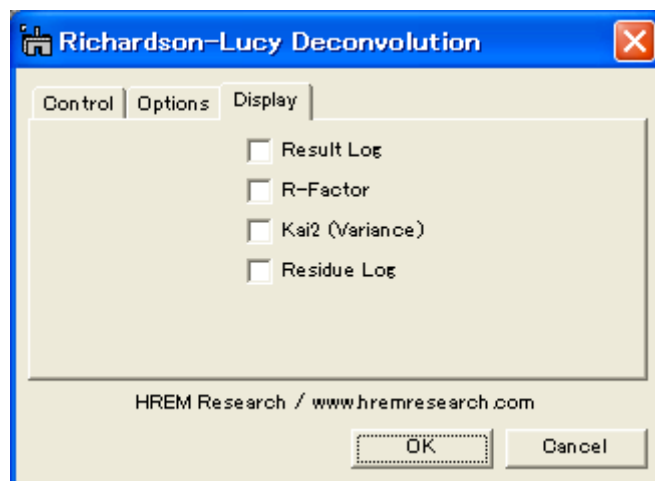
Convergence Factor: Actual modification of each cycle is an estimated change multiplied by this factor. Default value is one (1).

2.3.3 Display tab-panel

The last tab-panel is designed for selecting the various results to be displayed at the end of processing.

Figure 2-7

Richardson-Lucy Deconvolution (Display Tab)



Result Log: All the intermediate results specified by "Save Results at" will be saved.

R-Factor: History of R-factor.

χ^2 (Variance): History of squared sum of differences.

Residue Log: All the intermediate difference between an observed image and an estimated image (convolution between the probe and deconvolution) will be saved at specified cycles.

3. *Installation*

This chapter describes hardware and software requirements to run the DeConvEELS software and an installation procedure of the DeConvEELS plugin.

3.1 Requirements

The DeConvEELS runs under DigitalMicrograph environment, and the software and hardware requirements are similar to those for DigitalMicrograph.

3.1.1 Hardware requirement

The DeConvEELS requires a user key properly installed at a USB port. Therefore, you have to install a driver for the user key. Moreover, if you will to process a two-dimensional spectrum, you need an enough memory to store the ResultLog.

3.1.2 Software requirement

The following is a list of the software requirements necessary to run DeConvEELS:

- **DigitalMicrograph for Windows (GMS1.7.x or later)**
- **User Key Driver** (downloadable from the HREM web site)
- **IPU Plug-in** (downloadable from the HREM web site)

3.2 Software Installation

The following plug-ins should be placed in the folder “PlugIns” on the same level of the DigitalMicrograph for Windows:

- **DeConvBasicLib.gtk (DeConvBasicLib)**
- **DeConvEELS.gtk (DeConvEELS)**
- **DeConvEELS.dll (DeConvEELS.lib)**

The PlugIns folder should exist under a normal installation of the DigitalMicrograph.

DeConvEELS menu commands will be appeared under the EELS menu, when the DigitalMicrograph is launched after placing the plug-ins the PlugIns folder.

Note IPU plug-in should be installed to use DeConvEELS (see 3.4)

3.3 User Key Installation

Installing User Key Driver

NOTE: follow the procedure *without* inserting the USB key into an USB port.

Installing User Key Driver

1. Execute HASP HL GUI installer.
2. Continue installation by selecting 'Typical' from Setup type.

A driver corresponding to your Operation System will be installed.

Installing User Key

Simply plug your user key into an USB port. Your computer needs not to be restarted.

3.4 IPU Installation

Please download the IPU (Image Processing Utility) plug-in from the HREM web site and install it according to the ReadMe.txt file.

4. Getting Started...

This chapter briefly explains typical steps of the DeConvEELS. Before doing this section, you have to install the DeConvEELS plug-ins as well as the driver for your user key, and insert your user key.

4.1 Deconvolution

This section briefly describes steps of spectrum deconvolution using the Maximum Entropy method. Deconvolution using the Richardson-Lucy algorithm follows the same steps.

4.1.1 Open Spectrum

The deconvolution routine will work on open images. Therefore, you must open a spectrum to be restored and a low-loss spectrum as a kernel function.

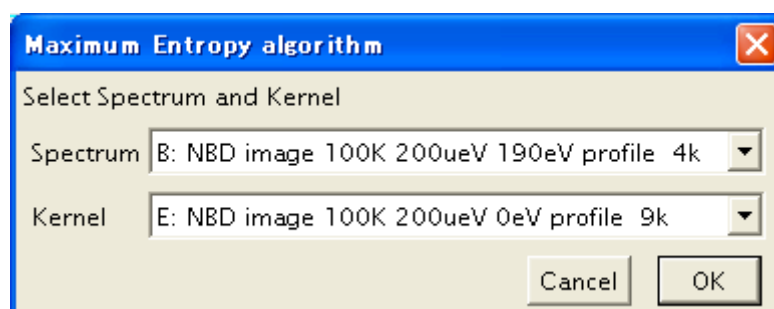
4.1.2 Launch Deconvolution

Select “Maximum Entropy” or “Richardson-Lucy” command from the Deconvolution menu as shown below. Here, we use the MEM as an example.



4.1.3 Select Spectrum

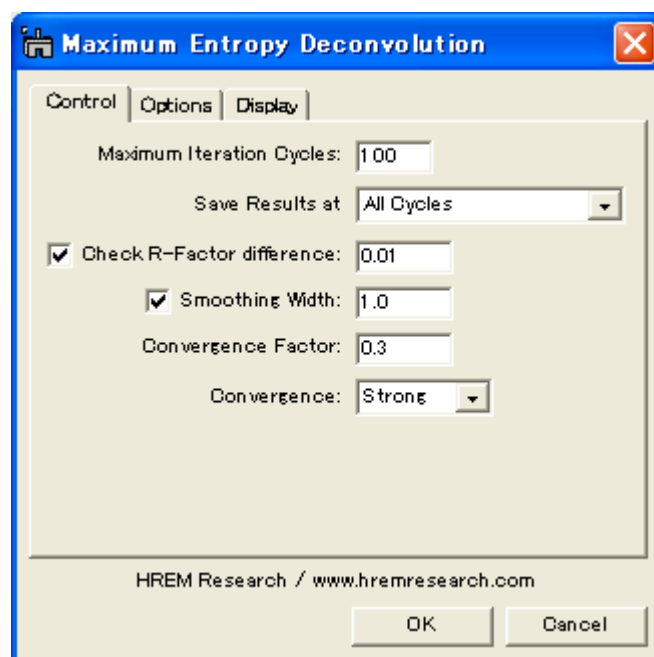
Select Spectrum and Kernel from the image list, and click OK to continue processing, or click Cancel to stop the procedure.



4.1.4 Setup Deconvolution Parameters

The deconvolution routine will work normally with defaults parameters as

shown here.

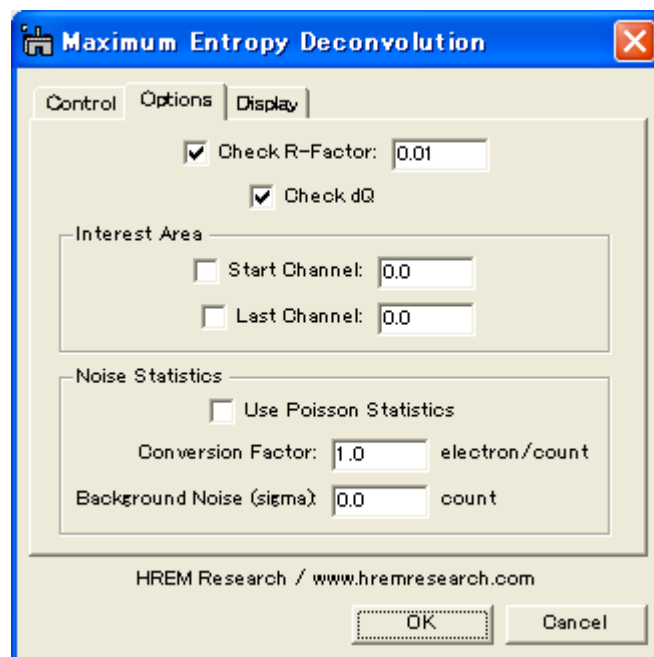


However, if your spectrum quality is too bad (noisy), or extremely good (high signal-to-noise ratio), you may change R-factor difference to a larger or a smaller value rather than the default value 0.01.

The default Convergence Factor is 0.3. If an iteration stop with the dQ criterion, you may use a smaller value.

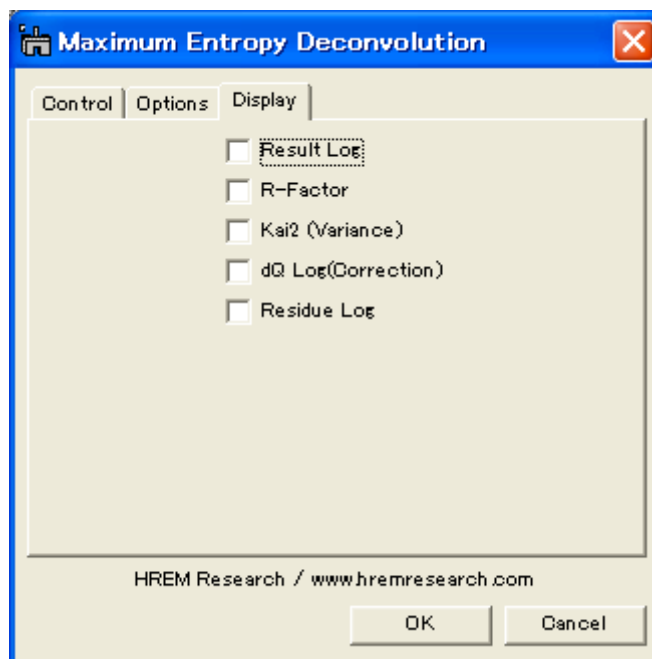
4.1.5 Check Optional Parameters

The following shows a typical setting for a core-loss processing. It is usually not necessary to neither specify Interest Area nor select Poisson Statistics for a core-loss processing.



4.1.6 Select Results to Display

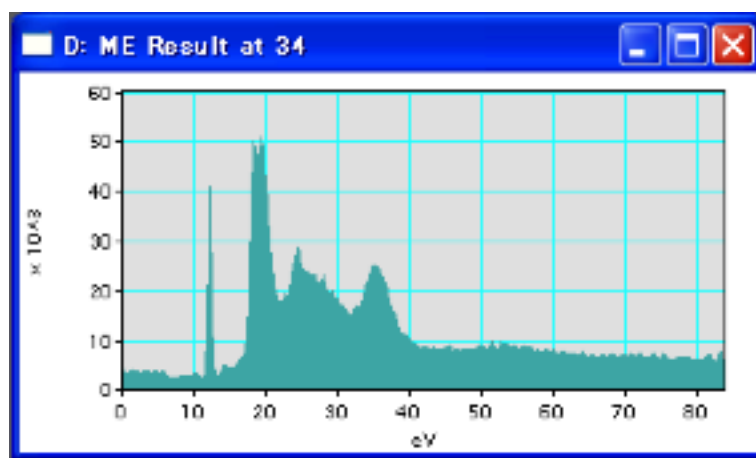
The final result after deconvolution will be always shown. If you check “Result Log” box, you can save all the intermediate results at specified cycles of deconvolution process, and check the results later by using a Line-Profile tool or a Slice tool. By selecting other three check boxes, you can see their histories.



4.1.7 Start Deconvolution

After setting conditions you can start deconvolution by clicking OK, or stop the procedure by clicking Cancel below the tab panels in the setup dialog.

Then, you will see a progress of deconvolution in the restored spectrum window as shown below:



4.1.8 Survey Results

The final result is displayed in the restored spectrum window as shown above. Some information on the progress of deconvolution will be shown in the DigitalMicrograph's Results Window.

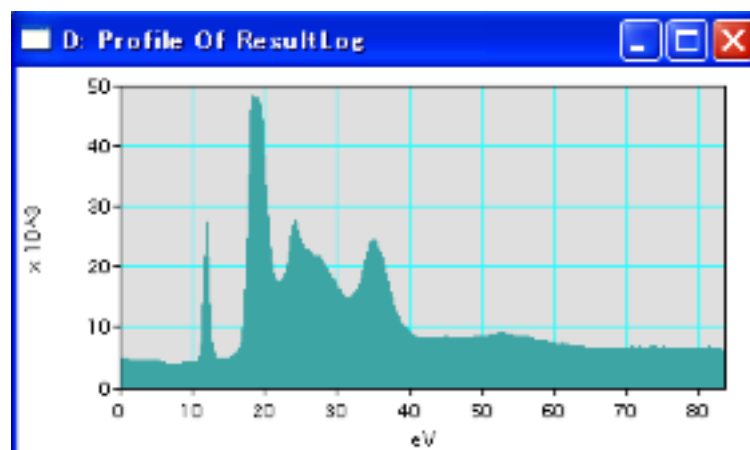
If you select "Result Log" check box in the setup dialog, you can save the intermediate results at specified cycles in the ResultLog.



The ResultLog image is easily surveyed by using a utility "Show as Movie" as explained later in the Utility section



By using the Line Profile tool in the Standard Tools palette, you can also check this result at any cycle as shown below.



4.1.9 SI-Richardson Lucy

Using this command you can process an EELS-SI data spectrum-by-spectrum using the Richardson-Lucy algorithm.

Before using this command, you have to process a single spectrum extracted from the SI data using the regular RL deconvolution command. In this way you can find the good process conditions before processing all the SI data spectrum-by-spectrum, which will take a substantial amount of time.

Important This command does not set-up process conditions. Therefore, you have to set-up the process conditions by processing a single spectrum extracted from the SI data.

Note The optimized SI deconvolution module based on both Maximum Entropy Method (MEM) and Richardson-Lucy algorithm (RLA) will be commercially available as an extension of the DeConvEELS (SI Module). Please send your inquiry to support@hremresearch.com.

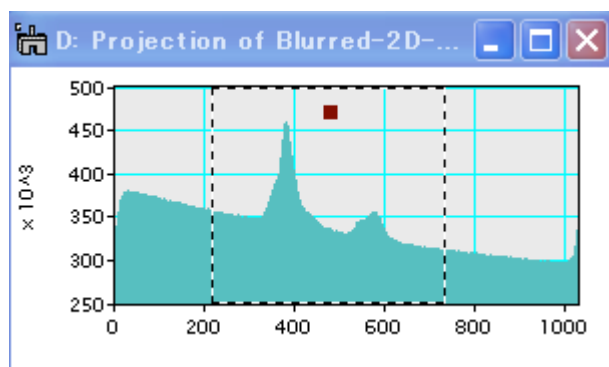
4.2 Selection and 2D Spectrum

4.2.1 Selection of Spectrum

Both deconvolution procedures work on a selection of spectrum of any size (not necessary to be a power of two).

A selection is created by using a range ROI (and its size is made a power of two by clicking the ALT-key when you drag a mouse).

Selection (Range ROI) on the Spectrum



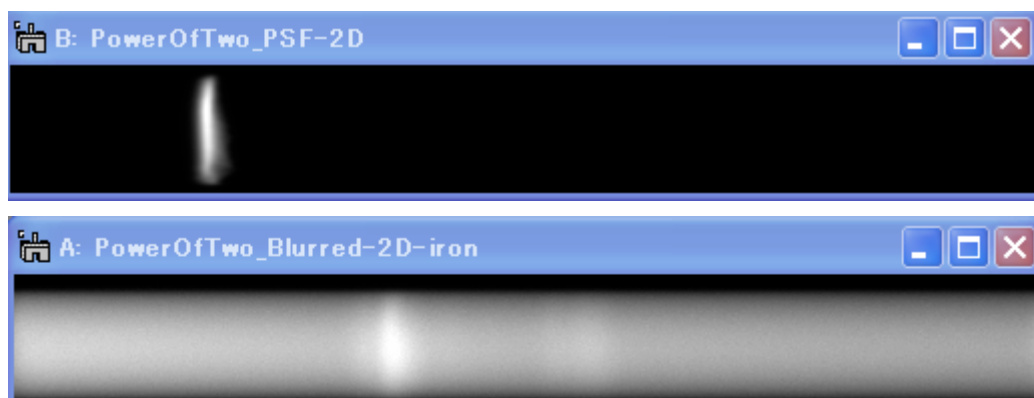
Note The sizes of the selected region of the kernel and the spectrum should be the same.

4.2.2 2D Spectrum

Both deconvolution procedures work on a selection of spectrum of any size (not necessary to be a power of two). As mentioned above, the procedures work also on a selection of 2D spectrum. The following shows an example of a 2D spectrum processing.

4.2.2.1 Sample 2D Spectrum

The following shows two sample 2D spectra of a core-loss region and a low-loss region.



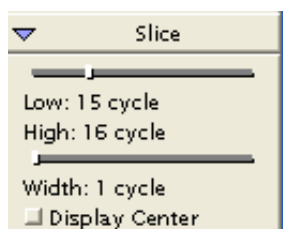
(These spectra were kindly provided by Dr. A. Gloter for testing the procedure. Please consult their original paper: Ultramicroscopy 96 (2003) 385-400)

4.2.2.2 Deconvoluted 2D Spectrum

The following shows a result obtained by the MEM procedure.



4.2.2.3 Survey your results



The ResultLog of a 2D image deconvolution is a 3D image, and easily surveyed by using a utility “Show as Movie” as explained later in the Utility section..

By using a Slice tool image as shown below, you can easily survey through the ResultLog image.

You can also check a deconvoluted spectrum at a selected cycle by extracting a slice (section) from the 3D image using “ Extract Slice” command in the “Utilities” sub-menu.

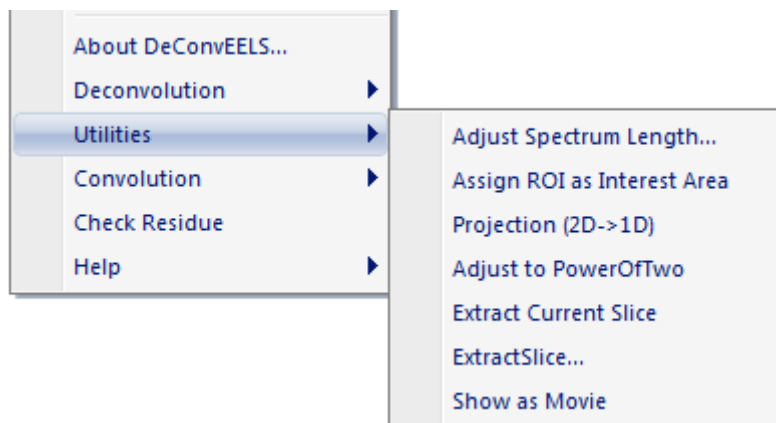


4.3 Utilities

Some useful commands to pre-process or post-process a spectrum are provided for your convenience.

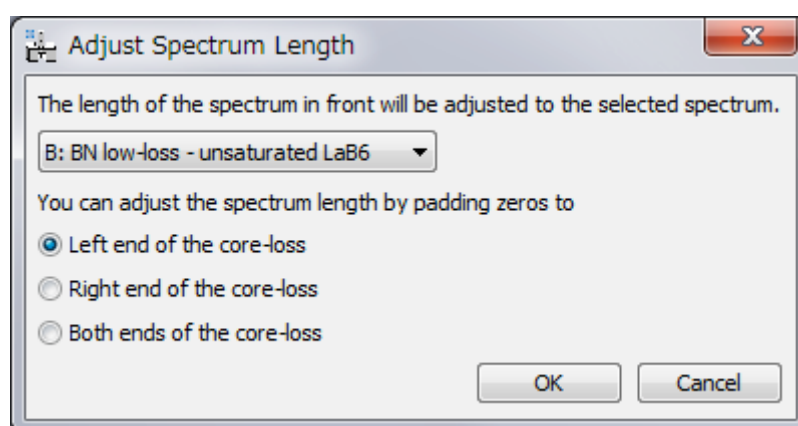
4.3.1 Utilities Menu

The following shows the "Utilities" sub-menu.



4.3.2 Adjust Spectrum Length...

The length of spectrum can be adjusted to the selected reference spectrum, which is normally a kernel (low-loss spectrum). This command is convenient to adjust the background subtracted core-loss signal, since the EELS command: Extract->Signal of GMS 2.x cuts the left-hand side of the background estimation region. You can adjust the spectrum length by adding zeros at the left side, or right side, both sides of the spectrum:



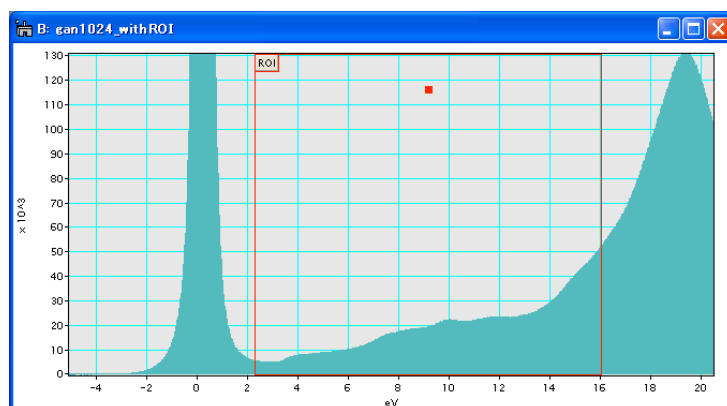
4.3.3 Assign ROI as Interest Area

A range ROI can be assigned as an Interest Area of the spectrum (see 2.2.2 and 2.3.2: Option tab). The interest area will be used to estimate convergence criteria. A convergence of the sharp feature like a zero-loss is slow, and affects

a convergence of the whole area. Therefore, the Interest Area is especially useful, if you want to process low-loss area (band-gap area) including the zero-loss.

How to use: Place a range ROI, and then use this command. Then the specified area changes to “ROI”. You can use this “Interest Area” ROI in conjunction with Spectrum Selection ROI (see 4.2.1)

Interest Area on the Spectrum



4.3.4 Projection Spectrum (2D->1D)

Deconvolution of a 1D spectrum is quicker than that of a 2D spectrum, and mathematically deconvolution of a projected data gives the same result as obtained by deconvolution of the original 2D data. Therefore, if you don't need to analyze an angular dependence of the spectrum, you may use 1D deconvolution. You can get a 1D spectrum from your observed 2D spectrum using this command. To use this command, you have to select a 2D spectrum before launching this command.

4.3.5 Adjust to Power of Two

Using this command you can adjust the size of your spectrum a selection of the spectrum to a power of two, which is required by the deconvolution routines. The size is adjusted to a next larger number that is a power of two. in order not to reduce resolution of the original image

To use this command, you have to select a spectrum before launching this command.

4.3.6 Extract Current Slice / Extract Slice...

Using these command you can extract a 2D spectrum (a slice) from the ResultLog (a 3D image) of a 2D deconvolution. Using **Extract Current Slice** command, you can extract the slice that is currently displayed. On the other hand, you will be asked to specify a slice number to be extracted, when you use **Extract Slice...**command.

To use these command, you have to select the ResultLog image before launching this command.

4.3.7 Show as Movie

Using this command you can survey through the ResultLog image like a movie. To use this command, you have to select the ResultLog image before launching this command. You can change an interval between the frames using the Left/Right Arrow keys, and change a direction (Forward/Backward) using the Up/Down Arrow keys. You can stop the movie by pressing the Space bar. This command will work for the ResultLog either from a 1D spectrum or a 2D spectrum..

4.4 Convolution

Two convolution routines are provided for your convenience.

4.4.1 Convolution Menu

The following shows the "Convolution" sub-menu.



4.4.2 Kernel

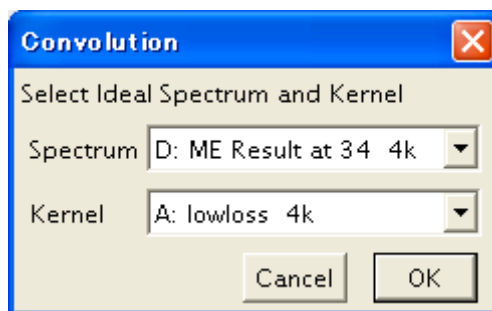
You may check your result by convoluting the restored spectrum and the kernel function (low-loss spectrum). The convolution should be close to the observed spectrum for a good deconvolution.

When you want to get a convolution for a final result, just select the final result, and next choose this command. Then, a result of convolution will be displayed. This is because the final result keeps information of the Kernel function.

When you want to get a convolution for other than the final result, you have to specify a kernel image. In this case the steps will be as follows:

1. Choose **Convolution** from EELS menu.

Then, the following windows to select images will appear. You have to open the images to be convoluted beforehand.



2. Choose Spectrum and Kernel images, and click **OK** to continue the process or click **Cancel** to stop the procedure.

A convoluted spectrum will appear in a minute.

4.4.3 Smoothing Gaussian

Using this command you can check a smoothed spectrum that will be used at deconvolution when you choose the Gaussian smoothing. When the smoothing widths specified for the MEM and RLA are different, you may be

asked the value you want to use. To use this command, you have to select a spectrum before launching this command.



4.5 Check Residue

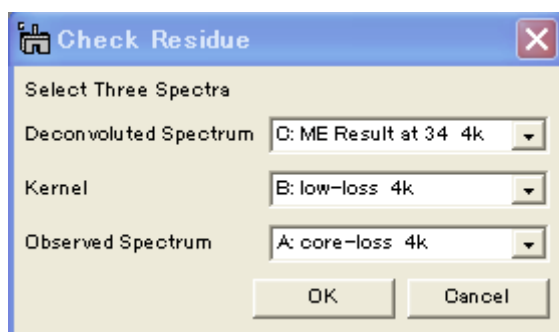
You can check residue for a result of deconvolution. The residue should be a random noise for a good deconvolution. Here, the residue means a difference between the observed spectrum and a convolution between the kernel and a deconvoluted spectrum. You can manually calculate the residue by subtracting a result obtained by the previous **Convolution** command from the observed spectrum. However, this is a handy command to calculate the residue.

When you want to get a residue for a final result, just select the final result and choose this command. Then, a residue will be displayed. This is because the final result keeps information of the original observed spectrum and the Kernel function.

When you want to get a result for other than the final result, you have to specify an observed spectrum and a kernel image. In this case the steps will be as follows:

1. Choose **Check Residue** from EELS menu.

Then, the following windows will appear to select images. You have to open three images to be used beforehand.



2. Choose **Deconvoluted Spectrum**, **Kernel** and **Observed Spectrum** then click **OK** to continue the process or click **Cancel** to stop the procedure.

A residue spectrum will appear in a minute.

5. Tips and Troubleshooting
