

Filtering Chromatic Aberration for Wide Acceptance Angle Electrostatic Lenses II. Experimental Evaluation and Software Based Imaging Energy Analyzer

Ádám Fazekas, Hiroshi Daimon, Hiroyuki Matsuda, László Tóth

Abstract—Here, the experimental results of the method of Filtering the effect of Chromatic Aberration (FECA) for Wide Acceptance Angle Electrostatic Lens (WAAEL) based system is described. This method can eliminate the effect of chromatic aberration from the images of a measured spectral image sequence by determining and removing the effect of higher and lower kinetic energy electrons on each different energy image, which leads to significant improvement of image and spectral quality. The method is based on the numerical solution of a large system of linear equations and equivalent with a multivariate strongly nonlinear deconvolution method. A matrix, which elements describes the strongly nonlinear chromatic aberration related transmission function of the lens system, acts on the vector of the ordered pixels of the distortion free spectral image sequence, and produce the vector of the ordered pixels of the measured spectral image sequence. Since the method can be applied not only on 2D real- and k-space diffraction images, but also along a third dimension of the image sequence, that sequence that is along the optical or in the 3D parameter space, the energy axis, it functions as a Software Based Imaging Energy Analyzer (SBIEA). It can also be applied in cases of light or other type of optics, for different optical aberrations and distortions. In case of electron optics the SBIEA method makes possible the spectral imaging without the application of any other energy filter. It is notable that this method also eliminates the disturbing background significantly in the present investigated case of Reflection Electron Energy Loss (REELS) spectra. It eliminates the instrumental effects and makes possible to measure the real physical processes better.

Keywords—Chromatic Aberration, Electron optics, Electrostatic lenses, Image sequences, Spectroscopy, Electron Trajectory

I. INTRODUCTION

Lens aberrations are fundamental and well known problems since the beginning of light and ion optics [1-3] that have strong effects on the performance of optical systems e.g. on the achievable angular-, spatial- and energy resolutions as well as on the contrast and information limits of the measured images. On the other hand, in the case of chromatic aberration, it can be useful to observe spectrally resolved images by using a small aperture at a focal- or imaging plane of a specific energy. Several decades have been required to find the practical solutions for these problems in cases of light and even more in electron and ion optics.

There are several methods in electron and ion optical aberration correction such as the correction of chromatic- and spherical aberrations by measuring the Time of Flight (TOF) of the imaging particles, but this method in the case of larger acceptance angle lens cannot distinguish whether a particle is arrived to the detector along a larger initial angle orbit and therefore a longer path, or just with smaller energy [4]. Wide Acceptance Angle Electrostatic Lenses (WAAEL) (Fig. 1) and WAAEL based Display-type Ellipsoidal Mesh Analyzers (DELMA) (Fig. 2) use a quasi-ellipsoidal shape mesh lens by which the spherical aberration can be corrected up to $\pm 60^\circ$ (1.00π sr) acceptance angle that can make possible higher transmission as well as larger angular region and therefore higher spatial resolution spectral imaging [5-16] compared to the earlier projection type electron optical instruments.

Fig. 1. The electrode arrangement and the paths of different energy electrons to demonstrate the effect of chromatic aberration (on the base of Fig. 1 of [8]) in the $\Theta = \pm 60^\circ$ (1.00π sr) WAAEL (Wide Acceptance Angle Electrostatic Lenses) (acceptance angle; $\Theta = \alpha_{\max}$).

However, the effect of chromatic aberration in first order is proportional to the electron energy that on the one hand makes possible the measurement of spectral images at different pass energies, but on the other hand it also has disadvantages especially by mixing the 3D angular-, spatial and spectral information (Figs. 1,3) [9,11,12,14,17,18]. It occurs by the phenomenon that the higher and lower kinetic energy electrons emitted from different points can also hit on the same pixels of the images of desired specific pass energy

Ádám Fazekas is with the Department of Informatics Systems and Networks at the University of Debrecen, Faculty of Informatics, Debrecen, Hungary (e-mail: fazekas.adam@inf.unideb.hu).

Hiroshi Daimon is with the Nara Institute of Science and Technology, Takayama, Ikoma, Nara, 630-0192 Japan (e-mail: daimon@ms.naist.jp).

Hiroyuki Matsuda is with the Nara Institute of Science and Technology, Takayama, Ikoma, Nara, 630-0192 Japan (e-mail: hmatsuda@ms.naist.jp).

László Tóth is with Department of Informatics Systems and Networks at the University of Debrecen, Faculty of Informatics, Debrecen and also with the SciTech MűszerKft., Debrecen, Hungary (e-mail: toth.laszlo@inf.unideb.hu, laszlo.toth@scitechmuszer.com).

(E_{pass}) electrons, which is the origin of blur and decrease of the sharpness as well as lowering the angular-, spatial and energy resolutions (Figs. 1,3) [9,14,17,18].

Fig. 2. The electrode arrangement and the paths of given E_{pass} energy electrons in the $\pm 45^\circ$ (0.59π sr) WAAEL based Display-type Ellipsoidal Mesh Analyzer (DELMA) (Fig. 11 of [15]).

Therefore, in the case of WAAEL based lenses the elimination of the distortion effect of the chromatic aberration is unavoidable to take the advantages that these types of electrostatic lenses provide.

Fig.3. (a) Schematic figure of the chromatic aberration in the case of a point source with three different kinetic energies. The solid trajectory line (E_{pass}) has a point image, but in the case of lower (dashed line, $E_{\text{imp}}=E_{\text{pass}}^2$) and higher (dotted line, $E_{\text{imp}}=E_{\text{pass}}^2$) impact kinetic energies, α -chromatic circles of confusion appear on the image plane with $r(x,y,\alpha,E_{\text{pass}},E_{\text{imp}})$ radii in α rad. The $\alpha=\angle AOB$ polar and $\beta=\angle COD$ azimuthal initial angles of the E_{pass} energy electron trajectory (solid line) are indicated. (b) The impact of chromatic aberration; from different sample points with different initial angles and kinetic energies produce overlapping circles of confusion, which degrades the image quality (Fig. 2 of [18]) and information content.

This problem also exists in the cases of simple-non-imaging electron spectroscopy (e.g. with Plain Mirror Analyzer (PMA), Cylindrical Deflection Analyzer (CDA), Cylindrical Mirror Analyzer (CMA), and others), where the high transmission and spectral resolution are the most important features, but not the imaging. [19]. ~~These methods use~~ uses an integrated beam of different energy electrons from a certain area of the sample that is projected onto the entrance slit of the spectrometer, which can be considered as a strongly distorted image. Therefore, the method of SBIEA can also be applied in those cases ~~for evaluating to eliminate~~ the instrumental effects.

Fig. 4. An example measured REELS spectral image sequence of a mesh sample (SUS 316, #100 line/in $\varnothing_{\text{wire}}=50\mu\text{m}$) (Tables 1, 6) excited by electron gun ($E_{\text{electron}}=1000\pm 0.45$ eV, $\varnothing_{\text{beam}}\approx 250\mu\text{m}$) (Tables 2) and measured by DELMA (in $E_{\text{electron}}=934-1032\text{eV}$ region with $\Delta E_{\text{electron}}=1\text{eV}$ steps) (Fig. 2, Tables 3, 4, 5) [15] furthermore, the spectra taken from the x, y coordinate pixels of ~~each image along~~ the measured spectral image sequence (straight line) and after the FECA SBIEA background subtraction procedure (dashed line).

In this or in cases of other spectral imaging systems the most commonly used image cleaning, ~~or~~ background elimination or deconvolution solutions methods act on a given energy image only, and do not take into account the energy and even not the dot by dot energy dependence of the lens aberrations' originated disturbing effects. A frequently used method in imaging and spectroscopy is a simple 2D deconvolution by a supposed Gaussian-type instrumental transmission function or an even more simple solution to get some image quality improvement, which divides the image by an averaged image sequence of the x and y direction shifted or rotated samples, in cases of real or angular imaging respectively [15]. An instrumental solution is given for this problem by the well-known, and in astronomy widely used idea of spectral ~~image~~ imaging spectroheliograph invented by G. E. Hale and H. A. Deslandres in the ~~first-third~~ decades of the 20th century [2019].

This method can also be applied in electron optics, where the WAAEL produced quasi monochromatic real- or angular image is projected to the entrance slit of e.g. a Concentric Hemispherical Analyzer (CHA). In this case the real- or k-space diffraction image is moved and therefore scanned in front of the entrance slit by the help of electrostatic or magnetic deflectors, while behind the exit slit of the CHA the selected energy spectral line, that is the monochromatic image of the entrance slit, is recorded and used to reconstruct the monochromatic image, slice by slice, parallel to the motion of the original image at the entrance slit [14,15].

We started to improve the quality of image data from the image processing side, in background fitting and subtraction for the integrated image intensities [14] and then the dot by dot Shirley-type background of the images were subtracted from the same coordinated pixels along the energy axes' of the spectral image sequences [17]. In this and the following cases all of the image points were taken into account as being very small entrance slits of independent spectrometers. This method resulted in quite good filtering close to the optical axes or in the case of Gaussian beams. The latest and also general method is the Filtering the Effect of Chromatic Aberration (FECA) for WAAEL [18] solves a system of linear equations (Eq. 1) where a vector (**b**), determined by the intensities of the image points of the aberrations and distortions free clear spectral image sequence is multiplied by a matrix (**A**) resulting the vector (**c**) of the intensities of the image points of the measured image sequence (Fig. 4). The matrix elements (a_{ij}) of **A** represent the effects of different energy spatial- or angular images' points on each other (Fig. 3), which can be determined by the numerical solution of the electrostatic Poisson- and relativistic electron motion equations that describes the physical behavior of the lens system. This strongly nonlinear 3D deconvolution-like method can enhance not only the real- and k-space image quality, sharpness, contrast as well as the position-, angular- and spectral information contents, but it can be applied as a Software Based Imaging Energy Analyzer (SBIEA). Furthermore, it is also possible to utilize the method to correct other, different types of aberrations [18].

The present paper is the sequel of the previous article [18], which discusses the theoretical concepts and model calculations of the filtering method on a simulated image sequence. Here, the ability of FECA method and its application as SBIEA are demonstrated on a spectral image sequence that have been measured by the High Voltage (HV) $\pm 45^\circ$ (0.59π sr) WAAEL based DELMA (Fig. 2.) [15]. In these cases significant image quality and spectral properties improvement have been observed by eliminating the electron backgrounds in the investigated cases of Reflection Electron Energy Loss (REELS) [219,224] and elastic electron spectra, only by taking into account the instrumental effects e.g. the chromatic aberration.

formázott: Alsó index

II. FILTERING THE EFFECT OF CHROMATIC ABERRATION (FECA) FOR WIDE ACCEPTANCE ANGLE ELECTROSTATIC LENSES (WAAEL) BASED DISPLAY-TYPE ELLIPSOIDAL MESH ANALYZER (DELMA)

Even with the latest electron optical systems the enhancement of the quality of the resulting images and spectra is a cardinal element of the measuring procedure. As it is shown above, there are several existing solutions to distinguish the disturbing background from the images with different approaches. However, none of them takes into account the complex behavior of the imaging system. Since in the case of WAAEL based lens systems (Fig. 1,2) [14,15] the spherical aberration has already been corrected [5-7], therefore in the present cases only the effects of the chromatic aberration is necessary to consider. Our intention was to develop a filtering method that provides a solution to filter the distortions of the spectral image sequences in both the image planes (x,y) and along the lens axes, or with other terminology energy (E) axes in the 3D parameter space. This method applies the solution of a system of linear equations (Eq. 1) where a matrix (**A**), whose elements describes the transmission function of the lens system, acts on the vector of the ordered intensity values of the pixels of the aberrations and distortions free spectral image sequence (**b**) resulting the vector of the ordered intensity values of the pixels of measured and aberration distorted spectral image sequence (**c**) (see ref. [18]).

$$\mathbf{Ab} = \mathbf{c} \quad (1)$$

The method of elimination of the effect of chromatic aberration previously was tested on a simulated image sequence and showed that the disturbing background can be removed sufficiently, while the useful information, which can be faded by the distortion effects, are enhanced making the future analysis of the measured data more feasible [18]. This process improves not only the image quality, but also the energy spectra taken from identically coordinated pixels (x,y) of the image sequence [18]. It was shown that the method can be improved further by considering the actual measurement environment more precisely, which means only the extension of the applied model and not requires the modification of the measuring instrument or any additional fabrication. So, since each pass energy (E_{pass}) of an optical system represents an aberration-disturbed image by the close and far different spatial positions, polar and azimuthal initial angles (α, β) to the sample surface (Fig. 3) and energy (E_{imp}) neighboring image points in the spectral image sequence, therefore if one can describe the optical, or in the present cases, electron optical behavior of the imaging system and can measure a series of images within a certain energy range with given energy differences (Fig. 4), then it becomes possible to identify and eliminate the distortion effect caused by different energy electrons originated from different positions of the sample (Figs. 3,4). In this way, e.g., the disturbing effect of the chromatic aberration can be modeled precisely in both real-, and k-space diffraction imaging and its effect can also be eliminated directly in cases of charged particles and other types of optics [18].

In terms of solvability, the key recognition was that the chromatic aberration filtering processes, which means the numerical solution of a strongly nonlinear Partial Differential Equation (PDE) system, can be separated into three independent parts. The first part requires electron optical calculations that is the numerical solution of the electrostatic Poisson- and then the relativistic electron motion PDEs and the determination of the electron trajectories leaving the sample from different positions (x,y), with different energies (E_{imp}), polar and azimuthal initial angles (α, β) to the sample surface (Fig. 3) that can affect the investigated E_{pass} electron energy image at the imaging plane. The second part is the determination of the transmission matrix (**A**) that describes the distortion effects of different energy and position image points of the image sequence to each other (Figs. 3,4) on the base of Eq. 2 and 3 (that is described in detail in ref. [18]) where the values of the matrix elements (a_{ij}), in the present case, are based on the determination of the chromatic "circle" of confusions for each point in the spectral image sequence (Eq. 2) by the help of the previously mentioned electron optical calculations. In other words each line in the **A** matrix represents a specific pixel in the image sequence with a given E_{pass} energy and x_{pass} and y_{pass} coordinates, and each entry in the line is the proportion of the intensity (Eq. 3) that arrived from the related pixel represented by the column ($E_{\text{imp}}, x_{\text{imp}}, y_{\text{imp}}$) to the pixel represented by the row ($E_{\text{pass}}, x_{\text{pass}}, y_{\text{pass}}$). This matrix represents the effect of chromatic aberration from a perspective of a single pixel. Finally, the third part is the iterative solution of the large system of linear equations, in the present case, by the relaxed Jacobi method with ω relaxation factor. The detailed description of the calculation is discussed in ChSec. 3 and 4 of ref. [18].

For simplification, in the present cases the resulted series of numerical data from the electron optical calculations are fitted and approximated by an $r(x,y,\alpha,E_{\text{pass}},E_{\text{imp}})$ polynomial that describe the radii of the chromatic circles of confusion (see Fig. 3) on the image taken at pass energy (E_{pass}) from a given impact energy (E_{imp}) and elevation angle (α) electrons, originated from different coordinates (x,y) of the imaged sample (Eq. 2, for detailed explanation see Eq. 1 of ref. [18]).

To accelerate the calculations further approximations were needed to be applied. Since the electron optical behavior of an electrostatic lens system remains the same in cases of proportional changes of the applied voltages or the geometry, therefore as a first approximation it was enough to make the numerical ray-tracing calculations and determine the $r(x,y,\alpha,E_{\text{pass}},E_{\text{imp}})$ polynomial only for a given $E_{\text{pass}}=E_0$ energy electrons (E_0) and electrode voltage configurations. In this case $E_0=1000\text{eV}$, the kinetic energy of the electron gun was chosen (The $\pm 0.45\text{eV}$ energy spread of the e. gun was not considered to simplify the calculation). The polynomials for different $\Delta E=E_{\text{imp}}-E_{\text{pass}}$ energy differences can then be determined with a simple multiplication by E_0/E_{pass} . To get a good approximation but keeping the simplicity of the equations, only the first order but different polynomials on the left ($E_{\text{imp}} \leq E_{\text{pass}}$) and on the right ($E_{\text{imp}} > E_{\text{pass}}$) side of the E_{pass} energy were applied (see Eq. (9) of Ref. [18]);

$$r(x, y, \alpha, E_{pass}, E_{imp}, E_e) = r(0, 0, \alpha, E_{pass}, E_{imp}, 1000 \text{ eV}) = r(\alpha, E_{pass}, E_{imp}) = \begin{cases} C_1 \alpha (E_{imp} - E_{pass}) \frac{1000}{E_{pass}} & E_{imp} \leq E_{pass} \\ C_2 \alpha (E_{imp} - E_{pass}) \frac{1000}{E_{pass}} & E_{imp} > E_{pass} \end{cases} \quad (2)$$

where C_1 and C_2 are the constant coefficients of the first order polynomials, which can be determined for a given $E_e = E_{pass}$ energy by the previously mentioned electron optical calculations. In our experiments, E_e was set to $E_e = 1000 \text{ eV}$. Furthermore, also for simplification, simplicity these first order polynomials were determined only at the optical axis (on-axes, $x=0, y=0$) and supposed to be the same for other off-axes image points (Eq.2). Although, this quasi linearity is in agreement with the general description of chromatic aberration [3], but in the future applications of FECA SBIEA methods, to get better and more accurate filtering, it is recommended to apply higher order and image position (x,y) dependent polynomials that will require much higher computation power. In this way the proportion of the intensity (I_a) of the E_{imp} energy electrons that comes from a given area of the sample, and goes through on the $\alpha + \Delta\alpha$ circular zone at the E_{imp} energy image of the spectral image sequence to the given pixel of the E_{pass} energy image of the spectral image sequence is given as (see Eq. 6 of ref. [18]):

$$I_a = T_{px} \frac{\cos(\alpha - \Delta\alpha) - \cos(\alpha)}{[1 - \cos(\theta)] \pi} \frac{r(\alpha, E_{pass}, E_{imp}, E_e)^2 - r(\alpha - \Delta\alpha, E_{pass}, E_{imp}, E_e)^2}{r(\alpha, E_{pass}, E_{imp}, E_e)^2} \quad (3)$$

where T_{px} is the area of an image pixel projected to and measured at E_{pass} energy at the detector and $\theta = \max(\alpha)$. That is for a given pass energy (E_{pass}), but different coordinated image point. The effect of a given impact energy (E_{imp}) and different coordinated (x,y) and angle (α) electron can be defined on the base of the $r(\alpha, E_{pass}, E_{imp})$ polynomial. Therefore, we can mark every pixel on the investigated energy image that has impact from a certain image point of a different energy (E_{imp}) image and calculate the proportion of its intensity that spreads on it. With this method a relationship can be defined between each point in the image sequence that describes the proportion of the intensity that impacts to other points of the sequence [18].

III. MAIN PARAMETERS FOR THE DERIVATION OF THE TRANSMISSION MATRIX ELEMENTS FROM NUMERICAL LENS SIMULATION CALCULATIONS AND THE MEASUREMENT ATTRIBUTES

In the present case the FECA SBIEA method [18] was applied on the spectral image sequence measured by the High Voltage (HV) $\pm 45^\circ$ (0.59 π sr) WAAEL based DELMA instrument (Figs. 2, 4) [15]. The spectral image sequence was taken from a SUS-316 metal alloy (Table 1:) [219] woven fine mesh sample. The sample was irradiated by an OCI-G10

electron gun (Table 2:) [232], the magnified images were intensified and converted into visible light by a double layered microchannel plate (MCP) plus, phosphor screen combined detector of the Hamamatsu Photonics (Table 3:) [243] and the images were recorded by a PCO Sencam QE thermoelectrically cooled CCD camera (Table 4:) [265].

The specific parameters of the FECA SBIEA calculations in the case of $\pm 45^\circ$ (0.59 π sr) WAAEL based DELMA lens is summarized in Tables 5-7. This lens system produced x12 magnification given pass energy (E_{pass}) real-space images on the entrance surface of the MCP at the last (4th) imaging plane. In this case the energy aperture at the 1st imaging plane was completely opened ($E_A, \phi_{EA} = 10 \text{ mm}$). The SUS-316 alloy (Table 1:) [219] #100 line/in $\phi_{wire} = 50 \mu\text{m}$ woven mesh sample was irradiated by $E_{e_gun} = 1000 \text{ eV}$ electrons with 16° inclination angle to the sample surface. The series of pass energies (E_{pass}) were set from 954 eV to 1034 eV with $\Delta E = 1 \text{ eV}$ steps by setting the electrode voltages (U1-4, L1-4, D1-3) (Fig. 2) [15]. The different pass energy (E_{pass}) images were recorded in 300x300 pixel resolution and 32 bit intensity depth that were reduced to 250x250 pixel sizes for the calculations.

Fig. 5. Montage of the measured spectral images of SUS 316 #100 line/in $\phi_{wire} = 0.05 \text{ mm}$ mesh sample (Table 1, 6), ~~excited by electron gun~~ ($E_{electron} = 1000 \pm 0.45 \text{ eV}$, $\phi_{beam} \approx 250 \mu\text{m}$) (Table 2) and measured by WAAEL based DELMA in $E_{electron} = 954-1034 \text{ eV}$ region with $\Delta E_{electron} = 1 \text{ eV}$ steps (Fig. 2, 4, Table 2-5, 7) [15]. The first and last images are copied $\times 10$ and added to the image sequence in lower and higher energy regions as constant linear boundary conditions (white frames) of the relaxed Jacobi method (Table 402).

Fig. 6. Montage of the FECA SBIEA filtered and stain reduced spectral images of SUS 316 #100 line/in $\phi_{wire} = 0.05 \text{ mm}$ mesh sample (Table 1, 6), ~~excited by electron gun~~ ($E_{electron} = 1000 \pm 0.45 \text{ eV}$, $\phi_{beam} \approx 250 \mu\text{m}$) (Table 2) and measured by WAAEL based DELMA in $E_{electron} = 954-1034 \text{ eV}$ region with $\Delta E_{electron} = 1 \text{ eV}$ steps (Fig. 2, 4, Table 2-5, 7) [15]. The white frames show the boundary conditions of the relaxed Jacobi method (Table 402).

The C_1 and C_2 constants of $r(x, y, \alpha, E_{pass}, E_{imp}, E_e)$ polynomial of Eq. 2 (Table 7:) that describes the ~~radiuses radii~~ of the chromatic circles of confusion on the measured images, had been primarily determined by numerical solution of the electrostatic Poisson-, and relativistic electron motion PDEs and by fitting the resulted discrete data series of the chromatic aberration dependent ~~radiuses radii~~ of the chromatic circles of confusion by first order polynomials around $E_e = E_{pass}$ energy. Then C_1 and C_2 were applied to determine the elements (a_{ij}) of the transmission matrix (A), in the way as it is described in ref. [18], but these constants (C_1, C_2) have to be refined by the feedback of the SBIEA calculations until the instabilities of the iterations have disappeared. The differences between the calculated and real values of C_1 and C_2 constants are originated from the calculation error as well as, the fabrication and composition tolerances furthermore, the accuracy and stability of the applied voltages etc. Also to avoid the instability of the iteration a boundary condition was necessary to be introduced to close the image sequence from lower and higher energy directions. For this reason the first and last images of the measured sequence were copied $\times 10$ and added to the sequence in lower and higher energy directions as a constant linear boundary condition. Finally, the handling of the screening effect of the energy aperture (EA) had to be

formázott: Alsó index

formázott: Alsó index

taken into account that additionally provided an approach to test the FECA SBIEA method.

IV. RESULTS

As it is described above the FECA SBIEA method [18] was applied on a spectral image sequence of electron gun irradiated (Table 2) metal alloy mesh sample (Tables 1,6,9) taken by $\pm 45^\circ$ (0.59π sr) WAAEL based DELMA (Fig. 2) (Tables 3-5) [15] electrostatic lens systems where the FECA SBIEA filtering algorithm [18] with given input parameters (Table 7) was implemented on the Integrated Supercomputer System of the National Information Infrastructure Development (NIIF), Hungary.

Fig. 7. The convergence of the relaxed Jacobi method in case of the WAAEL based DELMA images.

Fig. 8. The measured (a), FECA SBIEA filtered (b) and stain reduced filtered (c) spectral images of SUS 316 #100 line/in $\varnothing_{\text{wire}}=0.05\text{mm}$ mesh sample (Tables 1, 6) excited by electron gun ($E_{\text{electron}}=1000\pm 0.45\text{eV}$, $\varnothing_{\text{beam}}\approx 250\mu\text{m}$) (Table 2) and measured by the WAAEL based DELMA at the elastic peak ($E_{\text{electron}}=E_{\text{elastic}}=1000\text{eV}$) energy (Fig. 5, 6, Table 3, 4, 5) [15].

Fig. 9. The measured and stain reduced filtered images with the lines where the intensity distributions of Fig. 10 are measured as well as the 10 pixel diameter circular areas from where the spectral information of Fig. 11-13 are collected along the energy (E) axes of the spectral image sequence taken by the WAAEL based DELMA system at the elastic peak energy ($E_{\text{electron}}=E_{\text{elastic}}=E_{\text{pass}}=1000\text{eV}$).

The $\pm 45^\circ$ (0.59π sr) WAAEL based DELMA (Fig. 2) due to its acceptance angle, magnification (x12) and its application mode, produced good quality images (Fig. 5). In this case 80 pieces of different energy, 300×300 pixel resolution images were taken and reduced to 250×250 pixel size as well as the sequence was also extended by 10 copies of the first and last images as constant linear boundary conditions (Table 7). Therefore, in the case of the applied 100 pieces of reduced size images (Fig. 5) (c vector in Eq. 1) the number of elements of the transmission matrix (A) was $(6.25\times 10^6)^2=3.90625\times 10^{13}$. Then the solution of the large system of linear equations system (Eq.1) by the relaxed Jacobi method resulted the chromatic aberration free images (Fig. 6) (b vector in Eq. 1). The procedure required about 50-100 iterations (Fig. 7) and 50-100 hours running time. The $E_{\text{pass}} = 1000\text{eV}$ pass energy (at the elastic peak) measured and filtered monochromatic images are shown in figures 8 and 9, where the effect of the filtering is visible well. The background levels are also reduced significantly in all the three dimensions of the parameter space; in x, y (Fig. 10) and E (Figs. 11-13) showing in these cases the effectiveness of FECA SBIEA method.

Fig. 10. The relative intensity distributions of the measured and normed FECA SBIEA filtered and stain reduced images to the highest measured peak along the solid and dashed lines of Fig. 9 respectively.

Although, these are only the first experimental results of the FECA SBIEA method, it was able to eliminate the electron

background from the investigated Reflection Electron Energy Loss Spectra (REELS) and elastic electron peak of SUS-316 alloy mesh samples (Tables 1, 6, 9) [210,224] significantly (Figs. 11-13) and produced monochromatic images only by taking into account the instrumental effects without the application of any additional energy filter instrument, like CHA. Figures 11-13 shows REELS spectra of SUS 316, #100 line/in $\varnothing_{\text{wire}}=0.05\text{mm}$ mesh sample (Tables 1, 6), excited by an electron gun ($E_g=1000\pm 0.45\text{eV}$, $\varnothing_{\text{beam}}\approx 250\mu\text{m}$) (Table 2) and measured by the WAAEL based DELMA in $E=954\text{--}1034\text{eV}$ range with $\Delta E=1\text{eV}$ step size (Figs. 5, 6, Tables 2-5,7) [15], from different parts of the sample. The filtered spectra were normalized to the same elastic peak (1000eV) intensity as the measured so it is easier to compare them.

Fig. 11. Measured (solid line) and the normalized filtered (dashed line) FECA SBIEA filtered and stain reduced (dashed line) REELS spectra of SUS-316 #100 line/in $\varnothing_{\text{wire}}=0.05\text{mm}$ mesh sample (Table 1, 9) excited by electron gun ($E_{\text{electron}}=1000\pm 0.45\text{eV}$, $\varnothing_{\text{beam}}\approx 250\mu\text{m}$) (Table 2) and measured by WAAEL based DELMA in $E_{\text{electron}}=954\text{--}1034\text{eV}$ region with $\Delta E_{\text{electron}}=1\text{eV}$ steps (Fig. 5, 6, Table 2-5,7) [15] and collected from a 10px diameter mesh wire cross area (white circle on Figure 9) along the measured and the filtered image sequences respectively. The peaks are quite sharp due to the better energy resolution of the instrument were higher than the applied energy step-. It shows that not only the background level but the relative intensity of the peak is better on the filtered images, and the background rise up close to the edges because of the boundary conditions.

In these cases other generally applied methods like the deconvolution by a Gaussian-type instrumental function or the inelastic energy scattering related Shirley-, Tougaard-[254,276] and other type background correction procedures were not needed to be applied at all, or at least a lower degree as usual.

Fig. 12. Measured (solid line) and the normalized filtered (dashed line) FECA SBIEA filtered and stain reduced (dashed line) REELS spectra, of SUS-316 #100 line/in $\varnothing_{\text{wire}}=0.05\text{mm}$ mesh sample (Table 1, 9) excited by electron gun ($E_{\text{electron}}=1000\pm 0.45\text{eV}$, $\varnothing_{\text{beam}}\approx 250\mu\text{m}$) (Table 2) and measured by WAAEL based DELMA in $E_{\text{electron}}=954\text{--}1034\text{eV}$ region with $\Delta E_{\text{electron}}=1\text{eV}$ steps (Fig. 5, 6, Table 2-5, 7) [15] and collected from the five pieces of 10px diameter mesh wire cross areas (black and white circles on Figure 9) along the measured and the filtered image sequences respectively. On these summarized spectra the background reduction of the process is more visible. The background rises up close to the edges because of the boundary conditions.

Fig. 13. Measured (solid line) and the normalized filtered (dashed line) spectra, FECA SBIEA filtered and stain reduced (dashed line) REELS spectra of SUS-316 #100 line/in $\varnothing_{\text{wire}}=0.05\text{mm}$ mesh sample (Table 1, 9) excited by electron gun ($E_{\text{electron}}=1000\pm 0.45\text{eV}$, $\varnothing_{\text{beam}}\approx 250\mu\text{m}$) (Table 2) and measured by WAAEL based DELMA in $E_{\text{electron}}=954\text{--}1034\text{eV}$ region with $\Delta E_{\text{electron}}=1\text{eV}$ steps (Fig. 5, 6, Table 2-5, 7) [15] and collected from the five pieces of 10pixel diameter empty mesh hole areas (white crosses on Figure 9) along the measured and filtered image sequences respectively. The background rises up close to the edges because of the boundary conditions.

The resulted monochromatic images and also the spectra that belongs to different image points in these cases are not only sharper with better resolution, higher contrast and less background level, but closely free of the chromatic aberration related lens effects. Also the obtained information in the spatial (x,y), angular (α) and energy (E) dimensions of the spectral image sequence is supposed to be related more close

to the real physical conditions of the sample and show hidden spatial (and angular, but it was not investigated here) and spectral structures compared to the unfiltered measured images and spectra.

On the basis of the present results one can claim that the FECA SBIEA filtering method is capable to reduce the effect of chromatic aberration, but it can be possibly improved further by increasing the size of the measured energy region, raising the energy sampling rate (ΔE), application of nonlinear boundary condition and higher order off-axes $r(x,y,\alpha,E_{\text{pass}},E_{\text{imp}})$ polynomial for the better description of the electron optical system's behavior.

V. CONCLUSION

Lens aberrations are strong limiting factors of the spatial, angular and energy resolution of different lens systems from light to charged particle optics.

Here we investigated the elimination of the effect of chromatic aberration by a new method of ~~Filtering the Effect of Chromatic Aberration (FECA)~~ for ~~Wide-Acceptance-Angle Electrostatic Lenses (WAAEL)~~ [14] and WAAEL based ~~Display-type Ellipsoidal Mesh Analyzer (DELMA)~~ [15] as well as, applied that as a Software Based Imaging Energy Analyzer (SBIEA) on measured real-space image sequences [18].

The method is based on the numerical solution of a large system of linear equations where the inverse of a matrix (A^{-1}), whose elements represents the chromatic aberration dependent transmission functions of the lens system and based on electron optical calculations. This A^{-1} matrix acts on the vector (**c**) that contains the intensity values of image points of the measured spectral image sequence, which multiplication results in the vector of the chromatic aberration free intensity values of the cleaned images (**b**) and spectra dot by dots. Because of the method can clean the given pass energy 2D real- (x,y) and k-space angular (α,β) or with other words diffraction images and also works along the third energy (**E**) dimension in the parameter space, therefore it is functioning as ~~an Software Based Imaging Energy Analyzer (SBIEA)~~ [18] and can also be applied in cases of light or other optics and for other different aberrations' corrections too. In cases of filtering the effects of chromatic aberration and light optics the movement of the detector, while in cases of electron or charged particle optics the variation of the electric or magnetic fields are needed for taking a series of images with different pass energies (E_{pass}). For filtering other type of aberrations the questionable dependent parameters are needed to be defined, e.g. in the case of spherical aberration the series of images have to be taken with different acceptance angles or what is better with different acceptance angle zones.

The FECA SBIEA method showed good results in cases of the measured real-space image sequences taken by WAAEL based DELMA (see Figs. 2, 5-13) electron optical systems.

The FECA SBIEA method can eliminate the chromatic aberration originated background in significant degree from the measured image sequences and can increase their contrast, spatial resolution and signal-to-noise ratio. The elimination of this background increases the energy resolution pixel by pixel

along the energy axes such as many individual but cross-talking spectrometer with pixel size slits following each other (Figs. 5-13). This method can also be applied in cases of simple electron spectrometers too, whose main goal is only to form as small spot of the electron beam, originated from a certain area of the sample, at the plane of the entrance slit of the spectrometers, as possible. That spot, therefore can be taken into account as a bad quality image where then the effect of chromatic and other aberrations can also be eliminated by the FECA SBIEA and related methods.

These properties may open new directions in one way by eliminating the effect of chromatic aberration and in other way making possible the position and angle sensitive spectrometry as well as, spectral imaging without the application of additional energy analyzer like CHA.

In case of proper application of FECA SBIEA method the filtered real- and k-space diffraction images as well as, the related spectra contain only the sample related physical information. Furthermore, this FECA SBIEA method [18] can be applied for filtering other different type aberrations in cases of light- electron- and other optics.

VI. ACKNOWLEDGMENT

This work was partly supported by JSPS Grant-in-Aid for Scientific Research on Innovative Areas "3D Active-Site Science": Grant Number 26105001 and by the SciTech Műszer Kft. Debrecen, Hungary. Furthermore, we would like to thank to the National Information Infrastructure Development (NIIF) Program in Hungary for providing supercomputing facilities for the computations in the *emzskop* project.

REFERENCES

- [1] T. Mulvey, "Origins and historical development of the electron microscope," BRIT. J. APPL. PHYS., vol. 13, pp. 197-207, 1962.
- [2] R. P. Feynman, R. Leighton and M. Sands, "Ch. 29. The motion of charges in electric and magnetic fields, 29-5 The electron microscope," in The Feynman Lectures on Physics, Vols. II, Mainly electromagnetism and matter, Ch.29-5, Reading, Addison-Wesley, 1964.
- [3] J. Orloff, Handbook of Charged Particle Optics, CRC Press, 2009.
- [4] G. S. a. H. Spiecker, "Correction of chromatic and spherical aberration in electron microscopy utilizing the time structure of pulsed excitation sources," Journal of Vacuum Science & Technology B, vol. 20, pp. 2526-2534, 2002.
- [5] H. Matsuda, H. Daimon, M. Kato and M. Kudo, "Approach for simultaneous measurement of two-dimensional angular distribution of charged particles: Spherical aberration correction using an ellipsoidal mesh," Physical Review E, vol. 71, no. 6, pp. 066503-1-8, 2005.
- [6] H. Matsuda and H. Daimon, "Approach for simultaneous measurement of two-dimensional angular distribution of charged particles. II. Deceleration and focusing of wide-

- angle beams using a curved mesh lens," *Physical Review E*, vol. 74, no. 3, pp. 036501-1-9, 2006.
- [7] H. Matsuda, H. Daimon, L. Tóth and M. F., "Approach for simultaneous measurement of two-dimensional angular distribution of charged particles. III. Fine focusing of wide-angle beams in multiple lens systems," *Physical Review E*, vol. 75, no. 4, pp. 046402-1-5, 2007.
- [8] H. Daimon, H. Matsuda, L. Tóth and F. Matsui, "Stereo-PEEM for three-dimensional atomic and electronic structures of microscopic materials," *Surface Science*, vol. 601, no. 20, p. 4748–4753, 2007.
- [9] L. Tóth, H. Matsuda, T. Shimizu, F. Matsui and H. Daimon, "New Simple Photoemission Electron Microscope with an Energy Filter," *Journal of the Vacuum Society of Japan*, vol. 51, no. 3, p. 135–137, 2008.
- [10] L. Tóth, H. Matsuda and H. Daimon, "Simple method for making deeply curved mesh," *Journal of Electron Spectroscopy and Related Phenomena*, vol. 171, p. 64–67, 2009.
- [11] L. Tóth, K. Goto, H. Matsuda, F. Matsui and H. Daimon, "New 1π sr acceptance angle display-type ellipsoidal mesh analyzer for electron energy and two-dimensional angular distribution as well as imaging analysis," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 648, p. S58–S59, 2011.
- [12] K. Goto, H. Matsuda, M. Hashimoto, H. Nojiri, C. Sakai, F. Matsui, H. Daimon, L. Tóth and T. Matsushita, "Development of Display-type ellipsoidal mesh analyzer," *e-Journal of Surface Science and Nanotechnology*, vol. 9, pp. 311-314, 2011.
- [13] H. Daimon, H. Matsuda, F. Matsui, L. Tóth, M. Morita, S. Kitagawa and T. Matsushita, "Development of High-energy-resolution Display-type Photoelectron Spectrometer for Microanalysis," *Activity Report, Synchrotron Radiation Laboratory of the Institute for Solid State Physics, University of Tokyo*, Vols. (http://www.issp.u-tokyo.ac.jp/labs/sor/pdf/user/harima/daimon/AR2011_Daimon.pdf), 2011.
- [14] L. Tóth, H. Matsuda, F. Matsui, K. Goto and H. Daimon, "Details of 1π sr wide acceptance angle electrostatic lens for electron energy and two-dimensional angular distribution analysis combined with real space imaging," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 661, no. 1, p. 98–105, 2012.
- [15] H. Matsuda, K. Goto, L. Tóth, M. Morita, S. Kitagawa, F. Matsui, M. Hashimoto, C. Sakai, T. Matsushita and H. Daimon, "Development of display-type ellipsoidal mesh analyzer: Computational evaluation and experimental validation," *Journal of Electron Spectroscopy and Related Phenomena*, vol. 195, pp. 382-398, 2014.
- [16] H. Matsuda, L. Tóth, F. Matsui and H. Daimon, "Evaluation of disturbing effect of mesh holes in wide-acceptance-angle electrostatic mesh lenses," *Journal of Electron Spectroscopy and Related Phenomena*, vol. 195, p. 78–84, 2014.
- [17] Á. Fazekas, H. Daimon, H. Matsuda and L. Tóth, "Elimination of the Background of Electron Microscope Images by Using FPGA," *ActaCybernetica*, vol. 21, no. 1, p. 123–134, 2013.
- [18] Á. Fazekas and L. Tóth, "Filtering Chromatic Aberration for Wide Acceptance Angle Electrostatic Lenses," *IEEE Transactions on Image Processing*, vol. 23, no. 7, p. 2834–2841, 2014.
- [19] D. P. Woodruff and T. A. Delchar, *Modern techniques of surface science*, 2nd ed. Cambridge: New York: Cambridge University Press, 1994.
- [20] G. Hale, "The spectrohelioscope and its works. Part I. History, instruments, adjustments, and methods of observation," *The Astrophysical Journal*, vol. 70, no. 5, pp. 265-327, 1929.
- [21] "Stainless Steel - Grade 316L - Properties, Fabrication and Applications (UNS S31603)," 2015. [Online]. Available: <http://www.azom.com/article.aspx?ArticleID=2382>. [Accessed 31 03 2015].
- [22] W. Werner, K. Glantschnig and Ambrosch-Draxl, "Optical Constants and Inelastic Electron-Scattering Data for 17 Elemental Metals," *Journal of Physical Chemical Reference Data*, vol. 38, no. 4, pp. 1013-1092, 2009.
- [23] OCI Vacuum Microengineering, "Electron Guns G10 Series," [Online]. Available: http://www.ocvm.com/electron_guns_g10_series.html. [Accessed 22 11 2014].
- [24] Hamamatsu, MCP & MCP Assembly Selection Guide, Iwata: Hamamatsu Photonics K.K., Electron Tube Division, 2013.
- [25] D. A. Shirley, "High-Resolution X-Ray Photoemission Spectrum of the Valence Bands of Gold," *Phys. Rev. B*, vol. 5, no. 12, pp. 4707-, 1972.
- [26] PCO, "PCO Support, Senciscam QE," [Online]. Available: <http://www.pco.de/>. [Accessed 22 11 2014].
- [27] S. Tougaard, "Practical algorithm for background subtraction," *Surf. Sci.*, vol. 216, no. 3, pp. 343-360, 1981.

TABLES

TABLE 1. ELEMENTAL COMPOSITION OF SUS-316 METAL ALLOY MESH MATERIAL [219] FOR THE APPROXIMATE INTERPRETATION OF THE OBSERVED REELS SPECTRA (SEE FIG. 9, 16-18) [210-224]

Element	%
Fe:	63.995-70.995
Cr:	16.00-18.00
Ni:	10.00-14.00
Mn:	2.00-3.00
Si:	0.75
N	0.10
C:	0.08
P:	0.045
S:	0.03

TABLE 2, THE MAIN PARAMETERS OF THE APPLIED ELECTRON BEAM PRODUCED BY OCI-G10 ELECTRON GUN [232].

Applied beam energy	1000eV
Energy spread (given by the manufacturer)	$\pm 0.45\text{eV}$
Applied beam diameter	250 μm
Applied inclination angle of the electron beam to the sample surface	14°

Constant Intensity Boundary Regions at the left and right side ($E_{\text{pass}}^l - 10$) to E_{pass}^l and E_{pass}^l to ($E_{\text{pass}}^l + 10$) eV (see Fig. 10, 11, 15):
 \varnothing_{px} (pixel size of the measured images)

944-954 eV
1034-1044 eV
0.1087mm

TABLE 3, THE MAIN PARAMETERS OF F1094-9 DOUBLE LAYER MCP AND PHOSPHOR SCREEN COMBINED DETECTOR OF HAMAMATSU PHOTONICS [243].

Outer size:	24.8mm
Electrode area:	23.9mm
Effective area:	20mm
Channel diameter:	10 μm
Channel pitch:	12 μm

TABLE 4, THE MAIN PARAMETERS OF PCO SENSICAM QE CCD CAMERA OF PCO [265].

Resolution:	1376 \times 1040pixel
exposure time (t_{exp}):	
from	500ns
to	1000s
dynamic range	12bit

TABLE 5, Main parameters of the applied DELMA configuration (Fig. 4) [15].

Acceptance angle:	$\pm 45^\circ$ (0.59 πsr)
Magnification (in real-space imaging mode):	$\sim \times 12$
Energy Aperture diameter (\varnothing_{EA}) located at the 1 st imaging plane	10mm
Sizes of the measured images:	300 \times 300 pixel
Applied projectile electron energy ($E_{\text{projectile}}$):	1000eV
Electron beam inclination angle to the sample surface	16°

TABLE 6, Parameters of the applied mesh sample.

Mesh size:	#100 line/in
$\varnothing_{\text{wire}}$:	50 μm

TABLE 7, Main parameters of the FECA SBIEA calculations for DELMA.

Applied Acceptance Angle (AA) for calculation:	$\pm 45^\circ$ (0.59 π sr)
Applied Angle Step for the calculation ($\Delta\alpha$)	1°
Image size (reduced for the calculation):	250 \times 250px
Applied Pass Energy range (E_{pass}^l - E_{pass}^h):	954-1034 eV
Pass Energy step (ΔE_{pass}):	1eV
Relaxation factor of the Jacobi method(ω):	0.01
Constants of Eq. 2	
$C_{1\omega}$:	-0.007
$C_{2\omega}$:	+0.007