Measurements of the quantitative lateral analytical resolution at sputtered gold-layers with the FEG-EPMA JEOL JXA-8530F

D Berger and J Nissen
Technische Universität Berlin, Zentraleinrichtung Elektronenmikroskopie (ZELMI), Strasse des 17. Juni 135, 10623 Berlin, Germany
E-mail: dirk.berger@tu-berlin.de, joerg.nissen@tu-berlin.de

Abstract. In this paper we present measurements of the quantitative lateral analytical resolution of the field emission (FEG) electron microprobe JEOL JXA-8530F. In particular, the minimum thicknesses of gold layers that are suitable for an accurate quantitative analysis are determined for several measurement parameters. Both, low voltage analyses at acceleration voltages from 15 kV to 5 kV and low overvoltage analyses at 13 and 15 kV are regarded. For both methods a minimum analytical resolution well below 170 nm is obtained while the X-ray signal is 5 times lower for the low overvoltage method. Special care is taken for the preparation of the test specimen. A Ag-layer reduces fluorescence effects and the cleaved surfaces are polished by a gentle focused ion beam. The porosity of the gold layers is regarded by a separate measurement of the density (17.6 g/cm³). The results coincide well with Monte Carlo simulations of the source volume of generated X-rays if the reduced density of the gold layers and experimental determined values of the beam diameter are used. Additionally the results are used to investigate methods to estimate the width of the source volume of emitted X-rays by the superposition of the beam diameter and theoretical predictions of the width of the electron interaction volume. The linear sum of both gives a quite good agreement.

1. Introduction
Many attempts have already been made to determine and increase the lateral analytical resolution of electron probe microanalysers, this includes both the scanning electron microscopes (SEM) with energy-dispersive X-ray spectrometers (EDS) and the microprobes (EPMA) with wavelength-dispersive X-ray spectrometers (WDS). To review the obtained results, the following two criteria have to be regarded.

Firstly, it is necessary to distinguish between the qualitative and the quantitative lateral analytical resolution. While the first describes the size of the smallest particles or features that are recognized, the latter refers to the ability of an accurate quantification. In the latter case the particles or features have to be equal to or larger compared to the source volume of the emitted X-rays taking absorption and fluorescence effects into account. As pointed out by several authors it is additionally necessary to keep in mind that the size of this source volume is influenced not only by the interaction volume of the electrons inside the sample but also by the beam diameter of the electron probe [1-5]. The left part of figure 1 illustrates the interaction volume of electrons if a point-shaped probe is used. The right part of figure 1 shows the spread of the interaction volume by an increased probe diameter. The source
The volume of emitted X-rays changes accordingly. Actual Monte Carlo (MC) simulation programmes are able to consider the finite beam diameter [6]. Merlet and Llovet [1] proposed to consider this effect in theoretical treatments by the mathematical convolution of the beam diameter \(d_0\) and the classical range of X-ray generation \(R_x\) [19] reduced by depth of the maximum of the X-ray depth distribution \(Z_m\). Then the minimum analytical diameter \(R_S\) is given by:

\[
R_S = \sqrt{4 \cdot (R_x - Z_m)^2 + d_0^2}
\]  

in which \(R_x\) is calculated by the density \(\rho\), the atomic number \(Z\) and atomic weight \(A\), the primary electron energy \(E_0\) and the critical edge energy \(E_i\):

\[
R_x = \frac{33A}{\rho \cdot Z} \left( E_0^{1.7} - E_i^{1.7} \right)
\]

Both, MC simulations and theoretical predictions still have to be proven by measurements.

Secondly, there are two measurement strategies to increase the lateral resolution, the low overvoltage and the low voltage analysis [7, 8]. The low overvoltage method implies the use of an electron energy close to the critical ionisation energy of the chosen X-ray line. In most cases this will be energies of several keV for which the lens aberrations are small and hence the diameter of the electron probe is small and negligible in comparison to the interaction volume. The advantage of this method is that the electrons having enough energy to ionise the considered shell have undergone few scattering processes only. Therefore, the interaction volume of these electrons is small compared to the overall interaction volume for complete stopping of electrons. Then the source volumes of generated and emitted X-rays are small as well.

For the low overvoltage analyses, few values for the lateral analytical resolution have been published only and even then solely for the quantitative analysis. McSwiggen [7] estimated from MC simulations values of 200 nm and 150 nm for Fe-K\(\alpha\) and Ni-K\(\alpha\) respectively in a Fe50Ni50 compound. Berger and Nissen [4] have measured a resolution of 200 nm for Au-L\(\alpha\) at an Au-Ag-interface.

The low voltage analysis implies that the interaction volume of the electrons inside the sample is reduced by decreasing the electron beam energy well below 7 keV. As mentioned, the beam diameter has to be taken into account. It has been proven that the low voltage method is unusable for thermionic electron sources since the diameter of their electron beam often exceeds the interaction volume of the electrons [24]. Therefore, a field emission electron source is required for the low voltage analyses.
For the low voltage analyses many authors have published values for the obtained analytical resolution. The results for the qualitative lateral analytical resolution (table 1) range between 12 nm for W at 3 keV and 390 nm for Fe at 6 keV. Some authors use the recognition of small particles or layers as a measure for the qualitative resolution while others analyze the increase of the X-ray intensity across interfaces with an intensity criterion of 16% - 64% or 10% - 90%. In recent years the examination has been extended to specific problems that occur for the quantification in the low voltage method [3] especially due to the application of L-lines [9]. The few published values of the quantitative resolution (table 2) range between 115 nm for Si and 300 nm for Ti.

**Table 1.** Published values for the lateral resolution for qualitative X-ray microanalysis with the low voltage method.

| Element | Matrix | Electron energy [keV] | Qualitative lateral analytical resolution [nm] | Source |
|---------|--------|-----------------------|-----------------------------------------------|--------|
| Au      | Ni-Au-Cu interface | 5         | 70                                             | [10]   |
| Ni-layer semiconductor-layer | 3         | 31                                                | [11]   |
| W-layer | 3         | 25                                                | [12]   |
| Cu3Sn   | steel    | 6         | 100                                            | [13]   |
| Nb, C   | Sn       | 7         | 100                                            |        |
| Ag, Cu  |          | 8         | 100                                            |        |
| Ti      |          | 6         | 290                                            | [1]    |
| Fe      |          | 6         | 390                                            |        |
| cementite | steel   | ?         | 100                                            | [14]   |
| olivine |          | 7         | 250                                            | [15]   |
| orthopyroxene |          | 5         | 350                                            | [16]   |
| Au      | Si-Au interface | 5         | 26                                             | [4]    |
| Si      |          | 5         | 45                                             |        |

**Table 2.** Published values for the lateral resolution for quantitative X-ray microanalysis with the low voltage method.

| Element | Electron energy [keV] | Quantitative lateral analytical resolution [nm] | Source |
|---------|-----------------------|-----------------------------------------------|--------|
| Cu3Sn   | 6         | 300                                            | [13]   |
| Ag3Sn   |          | 250                                            | [17]   |
| Pd3Ga   | 5         | 750                                            | [18]   |
| Titanite | 7         | 300                                            | [15]   |
| MgO     | 7         | 300                                            |        |
| Cr      | 5         | 250 (Simulation)                               | [3]    |
| Ni      | 5         | 200                                            | [4]    |
| Au      | 5         | 120                                            |        |
| Si      | 5         | 115                                            |        |
Unfortunately, these published numbers do not give a satisfactory overview on the optimum quantitative resolution of a FEG-EPMA. Therefore, we started a systematic experimental determination of the quantitative lateral analytical resolution for different elements and different parameters, from which the first results for gold layers are presented in this paper. The resolution is determined by a standard-based quantitative point analysis in the layer. If the layer thickness is larger compared to the width of the source volume of emitted X-rays, then the sum of quantification is within (100.0 ± 0.5) %. In the next step the size of the layer is decreased until the sum is smaller than 100 %.

It is important to take absorption effects of the generated X-rays into account. Figure 2 shows Monte Carlo simulations (Casino v2.48 [6]) of the radial X-ray distribution with and without absorption. In the case of a primary electron energy of 5 keV the intensity of emitted X-rays is reduced at a maximum by 3 % with respect to the generated X-rays. For 15 keV the corresponding reduction is about 7 %. Therefore it is supposed that the difference of the widths of the source volumes of generated and emitted X-rays is negligible in our measurements.

![Figure 2. Monte Carlo simulations (Casino v2.48 [6]) of the radial X-ray distribution of Au-\(\alpha\) with (red) and without (blue) absorption for an Au specimen excited with 5 keV (left) and 15 keV (right) electrons.](image)

The focus of our investigations lies on the achievable resolution for an accurate quantification. All questions concerning signal intensity, precision and sensitivity are not regarded as a start. The experimental results are then compared to MC simulations and to predictions of theoretical considerations based on the ionisation range [1, 19, 20].

2. Experimental and results

2.1. Deposition and preparation of the gold layers

For the experimental determination of the quantitative analytical lateral resolution under different measurement conditions, test specimens were generated by sputtering gold layers with different thicknesses (1,000, 450, 335, 240, 200, 170 and 140 nm) on SIGRADUR glassy carbon substrates. The cross-sectional preparation of the specimen was done first by cleaving and then by polishing with a focussed Ga-ion beam (FEI Helios Nano Lab600 FIB). A gentle ion beam (30 kV, 90 pA) is used to avoid the deposition of Ga into the Au layer. The cross-section polishing process requires a protection layer consisting of Ga, Pt and C which is apparent on top of the specimen. To avoid fluorescence effects in the protection layer, all specimens were first covered with an intermediate Ag layer. Since the absorption coefficients of the considered Au-lines are nearly the same in Au and Ag and much
smaller in C (substrate) it is supposed that X-ray absorption effects in the adjacent layers can be neglected in our case.

Figure 3 shows high resolution secondary electron (SE) images of the cross-section of the 6 test specimens with the thinnest Au layer thicknesses. To take the obvious porosity of the sputtered layers into account for the quantification, we use a 1,000 nm sputtered Au layer with the same porosity as a reference standard. Backscattered electron (BSE) images were made as well to verify that there is no significant diffusion of Ag into the Au grains. Since the absorption of the Au-lines in Au and Ag is nearly the same, the resolution measurements are not affected by the Ag condensation on the walls of the Au pores.

![Cross-section images of test specimens with different gold layers (450 to 140 nm thickness)](image)

**Figure 3.** Cross-section images of the test specimens with different gold layers from 450 to 140 nm thickness.

For MC simulations the porosity is considered by a reduced density compared to that of bulk Au. The density of the layers is measured with the weighing method [21]. A Si wafer with size of 1 cm² is covered with an Au sputter layer whose thickness is measured to be 800 nm. The increase of the mass is determined with a precise balance and then the density is calculated to be $(17.6 \pm 1.1)$ g/cm³.

2.2. Measurements of beam diameter
As already mentioned and as illustrated in figure 1, the diameter of the electron probe has a significant influence on the lateral resolution of the quantification. Since all measurements are being made with the FEG-EPMA JEOL JXA-8530F, the beam diameters for different beam energies and beam currents are well known since they have been accurately measured. The results obtained from the derivative (DR) method specified in the standard ISO/TS 24597 [4] coincide well with those obtained by Fourier transform [3]. We use the beam diameter values from [4] that are derived from the average about 30,000 edge profiles of SE-images of gold insulars. To consider nearly all electrons for the
quantitative analytical resolution the edge profiles are analysed using the 1% - 99% criterion. The uncertainty of the beam diameters is expected to be less than 5%. The relevant beam diameters for the measuring parameters of this paper are summarized in table 3. The beam currents are chosen from a practical point of view to get a sufficient WDS-signal. As a rule thumb it can be said that the beam diameter is reduced by 30% if the current is reduced by a factor of 10. In most cases the resulting improvement of the analytical resolution is negligible since the electron range is much larger. But the reduced intensity prevents an accurate quantification.

| electron energy / beam current | beam diameter \(d_0\) [nm] |
|-------------------------------|------------------------|
| 3 keV / 1 nA                  | 52                     |
| 5 keV / 1 nA                  | 38                     |
| 8 keV / 1 nA                  | 31                     |
| 15 keV / 5 nA                 | 29                     |

2.3 Quantitative measurements

2.3.1. Low voltage method. Table 4 summarizes the results of the element quantification of the gold layers with the low voltage method using the Au-M\(\alpha\)-WDS-signal (2.12 keV, PET-crystal) at 15, 8 and 5 keV electron energy. Measurements were made in the “Spot Mode” using probe currents according to table 3 with 30 s measurement time for the peak and 10 s for the background. All measurements of table 4 and table 5 are repeated 5 to 12 times.

Using the 1,000 nm Au layer, no signal from the surrounding carbon and silver is detected even at 15 keV, therefore, it is applied as reference standard for the quantification (indicated by 100.00 in tables 4 and 5). A quantification result of (100.0 ± 0.5) % indicates that the source volume of the emitted X-rays is completely inside the Au-layer. For 15 keV solely the 1 \(\mu\)m layer can be resolved quantitatively, whereas for 8 keV a minimum layer thickness of 335 nm and for 5 keV a minimum layer thickness of even 170 nm is suitable for reliable quantitative analysis. Measurements with 3 keV were not successful due to the low intensity of the WDS-signal at 1 nA. Moreover, the accurate positioning of the electron beam is impossible due to the poor quality of SE-images (beam diameter about 50 nm at 3keV).

2.3.2. Low overvoltage method. The results of the low overvoltage quantitative measurements of the gold layers are summarized in table 5. Here, the Au-L\(\alpha\) X-ray line (9.71 keV, LiF-crystal) was used at 15 and 13 keV. All other parameters are the same. For 15 keV an accurate quantification is possible down to 335 nm layer thickness.

To optimize the lateral resolution in the low overvoltage condition, the electron energy was reduced to 13 keV, 1.08 keV more than the critical ionisation energy of Au L\(\alpha\) at 11.92 keV (overvoltage: 1.09). Indeed, now the 170 nm layer could be resolved quantitatively, comparable to the low voltage method. However, the X-ray intensity of Au-L\(\alpha\) reduces by a factor of 5, if the primary electron energy is decreased from 15 keV to 13 keV.
Table 4. Results of the quantitative measurements of the gold layers in wt% (low voltage method). The quantitative analytical lateral resolution is measured to be between 140 and 170 nm for 5 keV and between 240 and 335 nm for 8 keV.

| Au layer thickness [nm] | 15 keV (Au-Mα) | 8 keV (Au-Mα) | 5 keV (Au-Mα) |
|------------------------|----------------|----------------|----------------|
| 1,000 (Reference)      | 100.00         | 100.00         | 100.00         |
| 450                    | 92.04          | 99.93          | 99.89          |
| 335                    | /              | 99.81          | 99.82          |
| 240                    | /              | 88.08          | 99.48          |
| 200                    | /              | /              | 99.70          |
| 170                    | /              | /              | 99.71          |
| 140                    | /              | /              | 91.04          |

Table 5. Results of the quantitative measurements of the gold layers in wt% (low overvoltage method). The quantitative analytical lateral resolution is measured to be between 140 and 170 nm for an overvoltage of 1.09.

| Au layer thickness [nm] | 15 keV (Au-Lα) | 13 keV (Au-Lα) |
|------------------------|----------------|---------------|
| 1,000 (Reference)      | 100.00         | 100.00        |
| 450                    | 99.95          | 99.88         |
| 335                    | 99.76          | 99.83         |
| 240                    | 93.34          | 99.86         |
| 200                    | /              | 100.70        |
| 170                    | /              | 100.18        |
| 140                    | /              | 97.93         |

3. Discussion

3.1. MC simulations

3.1.1. MC simulations of the low voltage method. To estimate the source volume of emitted X-rays inside the gold, MC simulations of the interaction volume of the electrons inside the specimen were accomplished with 15, 8, 5 and 3 keV electron energy (figure 4), using the programme Casino (v2.48) [6]. The MC simulations of the electron paths in the sample are based on the reduced density of sputtered gold which was measured to be about 17.6 g/cm³. The uncertainty of about 6 % of the density measurements adds to the uncertainty of the MC simulations. All MC simulations incorporate the measured beam diameters. Therefore, the simulated interaction volume is expected to be a suitable estimation of the source volume of emitted X-rays and to the quantitative lateral analytical resolution. From MC simulations with and without absorption (figure 2) it has been proven that the absorption of the Au-lines in Au is negligible in the context of this work.
Following the simulations, the lateral diameter of the interaction volume for 99% of electrons decreases from about 550 nm at 15 keV to about 145 nm at 5 keV. A significant further decrease is not expected when the electron energy is further decreased to 3 keV, due to the increase of the beam diameter from 38 nm at 5 keV to 52 nm at 3 keV. At 5 keV, a decrease of the beam current by a factor of 10 would decrease the beam diameter by about 30% only. Therefore, the source volume would not change much but the x-ray intensity would decrease at the same time to an impractical value.

The simulations are in good agreement with the experimental data as can be seen from comparison with table 4.

3.1.2. **MC simulations of the source volume of generated X-ray lines in the low overvoltage method.**

The MC simulations in section 3.1.1 consider a complete stopping of the electrons inside the specimen. Therefore, these simulations are not suitable for the low overvoltage method for which the electron range of electrons with a certain energy loss matters only. This effect is considered by the MC programme PHI-RO-Z MONTE CARLO SIMULATION (v2.28) [22] that simulates the source volume of generated X-rays directly with respect to the energy loss of electron inside the sample. For our samples, the source volume of emitted X-rays is expected to be quite similar to the source volume of generated X-rays. This assumption is proved by the MC simulations of the radial X-ray distribution of emitted X-rays with and without absorption in figure 2. For our specimen and our measuring conditions the X-rays emission with absorption is at most 7% lower.
Figure 5 shows the results for Au-Mα (2.12 keV) and Au-Lα (9.71 keV) X-rays. The electron energy is 15 keV (left) and 13 keV (right) respectively. The lateral diameter of the source volume for 99% of X-rays decreases from about 500 nm for the Au Mα-line at 15 keV to about 140 nm for the Au Lα-line at 13 keV which is only 1.08 keV over the critical ionisation energy of Au-Lα (11.92 keV), resulting in an overvoltage of 1.09.

![Monte Carlo simulations](image)

**Figure 5.** Monte Carlo simulations (Phi-Ro-Z Monte Carlo Simulation v2.28) of the source volume of generated Au-Mα and Au-Lα X-ray lines at 15 and 13 keV electron energy.

Unfortunately, the beam diameter of 29 nm is not included in these simulations. But even without it, the simulations coincide very well with the measurements as well. Not only low voltage but also low overvoltage result in a significant enhancement of the lateral resolution.

3.2. Comparison to X-ray range calculations

Instead of simulating the individual electron paths inside the specimen, the effective ionisation range of electrons $R$ can be calculated [see paper by Fournelle et al. in these proceedings]. The most advanced formulae by Merlet and Llovet [1] consider the depth of the maximum of the X-ray depth distribution $Z_m$ and the beam diameter $d_0$ [1], see equation (1).

The value of $Z_m$ might be derived from MC simulations but are calculated by a formula from Merlet [23] here. The results from eq. (1) relevant to our measurements are summarized in table 6. The values of the range of X-ray generation $R_x$ have to be multiplied by a factor of 2 to compare to the resolution measurements from section 2.3. The factor of 2 is chosen because the range of X-ray generation $R_x$ describes the depth of the ionisation volume inside the sample and the width is expected to be the doubled value in Au, see figure 4.
Table 6. Calculated and measured values for the width of the ionisation volume and the lateral analytical resolution.

| Case     | Method       | \( Z_m \) from \([23]\) [nm] | \( 2 \, R_x \) [nm] Eq. (2) | \( R_S \) [nm] Eq. (1) | Exp. Result [nm] | \( d_W \) [nm] Eq. (3) | \( d_i \) [nm] Eq. (4) |
|----------|--------------|-------------------------------|-----------------------------|-------------------------|------------------|----------------------|----------------------|
| Au-M\(\alpha\) 5 keV | low voltage  | 6.9                           | 108.4                       | 101.8                   | 140-170          | 92.2                 | 146                  |
| Au-M\(\alpha\) 8 keV | low voltage  | 15.8                          | 284.6                       | 255                     | 240-335          | 173.3                | 315                  |
| Au-M\(\alpha\) 15 keV | conventional | 41.0                          | 897.6                       | 816                     | 450-1,000        | 477.8                | 920                  |
| Au-L\(\alpha\) 15 keV | low overvoltage | 7.6                           | 302.0                       | 287                     | 240-335          | 180.0                | 324                  |
| Au-L\(\alpha\) 13 keV | low overvoltage | 1.0                           | 100.4                       | 101.6                   | 140-170          | 79.2                 | 125                  |

Obviously, eqs. (1) and (2) calculate rather too small values for the high resolution conditions Au-M\(\alpha\) 5 keV and Au-L\(\alpha\) 13 keV. The discrepancy could be caused by wrong \( Z_m \) values or due to the fact that equation (2) has been derived for higher energies. Therefore, we will extend our measurements to layers consisting of further elements. The results will help to find the reasons for the discrepancies and to improve the formulae.

Willich and Bethke [24] proposed to calculate the lateral analytical resolution \( d_W \) by the sum of the range of X-ray generation \( R_x \) and the diameter width \( d_0 \) of the electron beam:

\[
d_W = R_x + d_0
\]

Unfortunately the obtained values are not in good agreement with the measured resolution probably because eq. (3) assumes that the width of the source volume of emitted X-rays is equal to the depth. But from the MC simulations in figure 4 it can be seen that the width of the electron scattering volume is about two times that of its depth. Therefore another attempt to calculate the lateral analytical resolution \( d_i \) is proposed that linearly adds the width \( d_0 \) of the electron beam and the estimated width \( 2 \, R_x \) of the source volume of emitted X-rays:

\[
d_i = 2 \cdot R_x + d_0
\]

From table 6 it can be seen that the values of (4) coincide well with the measurements, because this empirical formula considers better the shape of the interaction volume. Barkshire et al. [10] have observed and described this effect as well, finding a proportionality factor of 1.7 for Au, which obviously fits the measurements a little bit worse. However, eq. (4) will not be suitable for other elements than Au since the shape of the interaction volume depends on the density and probably on the primary energy. As soon as more measurements of the quantitative lateral analytical resolution of other elements are available, equation (4) has to be further developed.
4. Conclusions
In this paper it is experimentally proved that Au layers with thicknesses down to 170 nm are suitable for an accurate quantification in the EPMA. Therefore, the quantitative lateral analytical resolution is determined to be 170 nm for Au. This resolution is obtained with the low voltage method using 5 kV and the Au Mo-line as well as with the low overvoltage method using 13 kV and the Au La-line. The low voltage method requires an EPMA with a field emission source since the resolution is rather limited by the beam diameter than by the interaction volume of the electrons. A further decrease of the resolution for the low voltage method seems to be possible if a 3 kV-probe with a beam current well above 1 nA and a diameter well below 50 nm is available. For the low overvoltage methods no further improvements are to be expected since the resolution is independent from the probe properties and a further decrease of the overvoltage ratio decreases the X-ray intensity too strong.

For both methods the results of the quantitative lateral analytical resolution are in good agreement with MC simulations of the source volume of emitted X-rays. For the low voltage method it is mandatory to include the beam diameter in the simulation. Moreover, the achievable resolution can be estimated from measurements of the beam diameter and theoretical predictions of the range of X-ray generation. At least for Au, the linear sum of beam diameter and the doubled range of X-ray generation coincides well with the measured resolutions both in the low voltage and the low overvoltage method.

In the next step, the measurements of the quantitative lateral analytical resolution will be extended to further elements. Then experimental investigations of the quantitative depth-resolution in the EPMA will be of interest.

Acknowledgements
Financial support by the Europäische Fonds für Regionale Entwicklung (EFRE) within the project "Nano-Analytiklabor" is gratefully acknowledged. We thank Xavier Llovet for fruitful discussion and the support in applying eq. (1).

References
[1] Merlet C and Llovet X 2012 IOP Conf. Ser.: Mater. Sci. Engng. 32 012016
[2] Berger D and Nissen J 2012 JEOL User-Meeting (Göttingen, Germany)
[3] Pinard P T and Richter S 2014 IOP Conf. Ser.: Mater. Sci. Engng. 55 012016
[4] Berger D and Nissen J 2014 IOP Conf. Ser.: Mater. Sci. Engng. 55 012002
[5] Hombourger C and Outrequin M 2013 Microscopy Today 21 10
[6] Drouin D, Couture A R, Joly D, Tastet X, Aimez V and Gauvin R 2007 Scanning 29 92
[7] McSwiggen P 2014 IOP Conf. Ser.: Mater. Sci. Engng. 55 012009
[8] Newbury D 2002 J. Res. Nat. Inst. Stand. 107 605
[9] Statham P and Holland J 2014 IOP Conf. Ser.: Mater. Sci. Engng. 32 012016
[10] Barkshire I, Karduck P, Rehbach W P and Richter S 2000 Mikrochim. Acta 132 113
[11] Sakurada T, Hashimoto S, Tsuchiya Y, Tachibana S, Suzuki M and Shimizu K 2005 J. Surf. Anal. 12 118
[12] Hashimoto S, Sakurada T and Suzuki M 2008 J. Surf. Anal. 14 428
[13] Mori N 2010 JEOL News 45 42
[14] Bin W, Zhenyu L, Xiaoguang Z, Guodong W and Misra R D K 2013 Mater. Sci. Engng. A 575 189
[15] Armstrong J T, McSwiggen P and Nielsen C 2013 Microscopy and Analysis 27 17
[16] Saunders K, Buse B, Kilbrun M R, Kearns S and Blundy J 2014 Chem. Geol. 364 20
[17] McSwiggen P, Mori N, Takakura M and Nielsen C 2012 Microsc. Microanal. 17 Supp. S2 624
[18] Müller D, Schwerin J, Gille P and Fehr K T 2014 IOP Conf. Ser.: Mater. Sci. Engng. 55 01201
[19] Castaing R 1960 Adv. Electron. El. Phys. 13 317
[20] Rinaldi R and Llovet X 2015 Microsc. Microanal. 21 1053
[21] Berger D and Galbert F 2011 Conf. Proceedings MC2011 (Kiel, Germany) ISBN 978-3-00-033910-3 IM7.P208

[22] Joy D 1995 Monte Carlo Modeling for Electron Microscopy and Microanalysis (Oxford: Oxford University Press)

[23] Merlet C 1994 Mikrochim. Acta 114/115 363

[24] Willich P and Bethke R 1996 Mikrochim Acta Suppl. 13 631