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Paper

Determination of Surface-Excitation Parameters for Elastic Peak Electron Spectroscopy (EPES) Using the Database of Goto

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The inelastic mean free path (IMFP) can be determined from the elastic-backscattering probability I_e of electrons that can be measured by elastic peak electron spectroscopy (EPES). We calculated IMFPs from the absolute I_e measurements of Goto. Calculated values of the elastic backscattering-probability I_{ec} can be obtained from the EPESWIN software of Jablonski. I_{ec} denotes the number of electrons/incident electron, backscattered elastically and detected with the cylindrical mirror analyzer (CMA) of Goto. The I_e data deduced from the database of Goto are lower by 20-34% than the calculated I_{ec} for Si, Ni, Cu, Ag and Au. This discrepancy is explained by surface-excitation losses characterized by the SEP parameter. We made corrections for the experimental I_e data for surface excitations using the material parameters of Chen (modified), Kwei, Ding, and Werner. The parameters of Chen were modified for achieving the minimum deviations between the calculated of I_{ec} and the SEP corrected $f_s I_e$ values. The material parameter of Nagatomi a_{ch} = 4.3 was confirmed for Ni by EPES. The SEP corrected IMFPs were deduced from the SEP corrected $f_s I_e$ data. SEP correction of the IMFPs resulted in mean deviations from the calculated TPP-2M data of 3.9 % for Si, 6-7% for Ni, 8.2% for Ag and nearly 12 % for Cu and for Au.

1. Introduction

The inelastic mean free path (IMFP) of electrons can be determined experimentally by elastic peak electron spectroscopy (EPES) [1,2,3]. The IMFP is calculated from the elastic-backscattering probability I_e, which is derived from the measured elastic-peak intensity. This calculation was made with the EPESWIN software of Jablonski [4] which is based on the NIST SRD 64 [5] and SRD 71 databases [6]. The IMFP is calculated with the TPP-2M formula [7]. Comparison of the experimental I_e probability with the corresponding calculated I_{ec} shows that: $I_e < I_{ec}$. This difference is due to surface excitation losses, characterized by the surface excitation probability, the (SEP) parameter P_{se}. It is defined by ISO [3] as "for AES, EPES, REELS, XPS, the characteristic parameter in the exponential attenuation, describing the ratio of the intensity of a peak resulting from the presence of the surface, during a single crossing of a material surface, to that expected after traversing the same amount of material, but in absence of the surface". Assuming a Poissonian distribution of surface excitations, the elastic-peak intensity is reduced by $\exp(-P_{se})$. Tanuma *et al.* [8] introduced this definition. For evaluating EPES experiments, the experimental I_e is needed in absolute units, whereas the EPESWIN software supplies the calculated I_{ec} elastic-backscattering probability [4]. The measurement of $I_e(E)$, a function of electron energy E, in absolute units is very difficult [1].

The elastic current $i_e(E)$ is measured with the special CMA spectrometer of Goto [9]. The primary current i_p is kept constant and measured with a Faraday cage mounted on the sample holder. Goto's $i_e(E)$ experimental current is detected also with a Faraday cage. His data for the total backscattered spectra are available on the internet [10] for many elements, covering the energy range E = 50 eV – 5 keV and angular parameters for incidence α_i

= 0°, α_d = 42.3±6° (detection angle with respect to the surface normal). The determination of the transmission $T_r(E)$ of the CMA is hard but has been was determined experimentally by Goto *et al.* [11]. Very recently the backscattering yield (BY) for 10 elements [12] was used as reference for EPES. BY is defined by ISO [13]. $T_r(E)$ was deduced from BY [14] and resulted in reasonable agreement with Goto's experimental data [11].

2. Experimental procedure for determining the SEP parameter

Very recently, Tanuma *et al.* deduced the IMFP [15] for several elements from the elastic current $i_e(E)$ data of Goto. They did not make a correction for surface excitations. The experimental elastic backscattering probability $I_e(E)$ for the angular conditions of Goto is given by:

$$I_e(E) = \frac{i_e(E)}{i_p(E)T_r(E)} \tag{1}$$

with notations $i_e(E)$ the measured elastic current, i_p = 1 μA the primary beam current [10] provided that the energy resolution of the CMA, $\Delta E_s(E) = 0.0025E > \Delta E_e$, the full width at half- maximum (FWHM) of the elastic peak. The physical FWHM of the elastic peak is determined principally by the FWHM of the electron gun [1]. For the CMA of Goto this condition is fulfilled for E > 500 eV. The elastic current i_e is an integral of the physical elastic peak. At the low energy limit $E_{e,min}$ of the experimental elastic peak, however, the energy-loss intensity is included in the vicinity of $E_{e,min}$ due to the energy resolution of the CMA. In our present work, a background correction [16] was applied for $i_e(E)$. From the experimental $i_e(E)$ data, uncorrected IMFPs λ_u were presented by Tanuma [15].

The present paper deals with the SEP correction of the $I_e(E)$ data of Goto. I_e was corrected for 5 elements: Ni, Cu, Si, Ag and Au, covering the energy range E = 0.4-1.5 keV. The SEP parameters were estimated, applying the model of Chen [17]. In addition, the material parameters of Kwei [18], of Ding [19] and of Werner [20] were tested.

The I_{ec} elastic-backscattering probability values were calculated with the EPESWIN software for E = 0.4, 0.5, 0.6, 0.8, 1, 1.2 and 1.5 keV [4]. The IMFPs λ_i were taken from NIST SRD 71 [6]. According to Tanuma *et al.* [8], the correction of I_e(E) for surface excitations is:

$$\frac{i_e}{\exp(-P_{se})} = I_{ec} \tag{2}$$

with the notations: I_e experimental probability, I_{ec} calculated probability and P_{se} the SEP parameter. The correction factor f_s = 1/exp(-P_{se}). Perfect correction would result in $I_{ec} = I_e f_s$, but this was never achieved. The ΔI_e average deviation between the experimental $I_e(E)$ data and the calculated $I_{ec}(E)$ data was found:

$$\Delta I_e\ \% = 100\frac{1}{n}\sum_{i=1}^{n}\left|\frac{I_e - I_{ec}}{I_{ec}}\right| \tag{3}$$

A similar equation is used for the SEP corrected $I_e f_s$ elastic-peak intensities with different SEP parameters. They are: f_{sC} (Chen modified), f_{sK} (Kwei), f_{sD} (Ding), f_{sW} (Werner). The efficiency of SEP correction is characterized by Eq. (3) applied for the selected f_s factor and the products $f_s I_e$.

The SEP parameter is given by the sum of probabilities of surface losses for incidence and escape, respectively:

$$P_{se} = P_{se,incident} + P_{se,escape} \tag{4}$$

$P_{se,incidence}$ and $P_{se,escape}$ can be described with the relationships used by Chen [17], Kwei [18], Ding [19] and Werner [20]:

$$P_{se,incident} = aE^{-b}\left(\cos\alpha_j\right)^{-c} \tag{5}$$

α_i = angle of incidence. A similar relationship is valid for escape. a, b and c are material parameters, different for incidence and escape. Chen [17] applied only one material parameter a_{ch}, and chose b=0.5 and c =1 values, identical for incidence and escape. Kwei et al. [18] and Ding *et al.* [19] applied 6 parameters. Werner *et al.* [20] applied the relationship of Oswald [21]. They revised their earlier work of 1999, cited in [20]. They obtained new, improved material parameters from recent REELS experimental results, using the Oswald formula:

$$P_{se} = \frac{1}{0.171a_W\sqrt{E}\cos\alpha + 1} \tag{6}$$

Very recently Jablonski and Zemek evaluated AREPES (angular resolved) experimental results [22]. They studied the elastic-peak intensity versus the angle

of detection α_d, working with a hemispherical analyzer (HAS) and normal incidence. They calculated the elastic peak intensity variation with α_d, with and without SEP correction. They checked the Chen [17] and Oswald models [21] for P_{se}(E), using Eq. (5) and Eq. (6) of Oswald. SEP correction for AREPES will be published in a forthcoming paper.

In our previous work [23], the material parameters of Chen [17] were applied for the elastic peak intensity ratios of Si/Cu, Si/Ni, Si/Ag and Si/Au. Since a_{chNi} was not published by Chen, we determined it by trial and error a_{chNi} = 3.6. Very recently this value was confirmed by Nagatomi and Goto [24] working with REELS spectra of Goto. In the present work, we modified the material parameters of Chen [17] by applying a trial and error method, for obtaining the best SEP correction of Goto's data [10], with minimum deviations between I_e, $f_{sC}I_e$ and I_{ec}, using Eq. (3). Nagatomi verified the model of Chen and found a_{chNi} =4.3. This value agreed reasonably with our value of 3.6.

The experimental $i_e(E)$ data of Goto [10] for Si, Ni, Cu, Ag and Au have been corrected for surface excitations. They are used as reference samples for electron spectroscopy and surface analysis [1,2]. The material parameters of Chen [17] were modified as described above. The material parameters f_{sK} of Kwei [18], f_{sD} of Ding [19] and f_{sW} of Werner [20] have been tested using Eq. (3). The uncorrected I_e curves are compared with I_{ec} calculated ones and with the corrected $I_e f_s$, using parameters of Chen (modified), Kwei, Ding and Werner. The material parameters are summarized in Table 1.

The uncorrected $I_e(E)$, the SEP-corrected $I_e f_s$ and the calculated $I_{ec}(E)$ results are displayed for Si in Fig. 1, and for Ni in Fig. 2. The other elements are similar. The results are summarized in Table 2.

3. Correction of the IMFP for surface excitation losses

The goal of the present work is the improvement of the experimental IMFP by the surface excitation correction. For determining the uncorrected λ_u IMFPs by EPES, the elastic backscattering probabilities $I_e(E)$ have been deduced from the $i_e(E)$ data of Goto [10]. The EPESWIN software is suitable for determining the uncorrected λ_u IMFPs.

The experimental determination of P_{se} according to Tanuma and the new ISO definition [3] needs the measurement of the elastic peak in absolute units. Goto measured the current $i(E)$ of the backscattering spectra $N(E)$ and of the elastic peak for i_p = 1 μA incident beam current. $i(E)$ refers to the backscattered electrons detected

Table 1. Material parameters applied for SEP-correction. Notations: a_i, b_i, c_i (incidence), a_o, b_o, c_o (detection).

		element				
		Si	Ni	Cu	Ag	Au
Chen original [7] Eq. (5)		2.5	-	2.45	2.34	3.06
Chen modified Eq. (5) [present work]		3.2	4.3	4.0	4.5	3.2
Werner Eq. (6) [20]		1.2	1	-	2.3	-
Salma-Ding Eq. (5) [19]	a_i	-	1.71	1.90	2.58	2.16
	b_i	-	0.44	0.40	0.44	0.44
	a_o	-	1.23	1.39	1.82	1.64
	b_o	-	0.37	0.46	0.37	0.39
Kwei *et al.*, Eq. (5) [18]	a_i	1.13	1.424	1.3	1.426	1.884
	b_i	0.476	0.476	0.466	0.482	0.506
	c_i	1.05	1.12	1.113	1.05	1.06
	a_o	1.73	2.17	1.999	2.12	1.87
	b_o	0.42	0.428	0.417	0.426	0.405
	c_o	0.83	0.79	0.82	0.74	0.8

within ΔE_s energy resolution. $N(E)$ denotes the number of backscattered electrons for E and energy steps ΔE = 1 eV. The experimental $i(E)$ data are affected by the energy resolution of the CMA $\Delta E_s = 0.0025E$ (eV) and thus:

$$N(E) = \frac{10^{-3} i(E)}{\Delta E_s(E) T_r(E)} \quad (7)$$

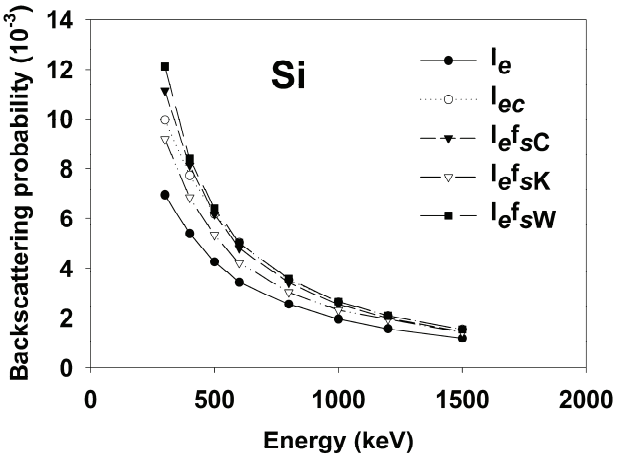

Fig. 1. Comparison of the elastic- backscattering probabilities versus E for Si. Notations: I_e uncorrected (deduced from Goto's database), $I_e f_{sC}$ SEP corrected with Chen, $I_e f_{sK}$ with Kwei and $I_e f_{sW}$ with Werner. I_{ec} calculated by EPESWIN.

The transmission $T_r(E)$ of the CMA of Goto [11] however, was problematic since it was based on optical measurements. Recently Goto determined $T_r(E)$ with an electron gun and electrons incident on the CMA [11]. It is slightly changed above 500 eV. In our recent work, quantification of the elastic peak was based on the backscattering yield (BY), calculated by L Zommer [12] and determined experimentally by integrating the $N(E)$ spectra [14]. Zommer achieved good agreement with experimental BY data for 10 elements (e.g. Si, Ni, Cu, Ag, Au etc), obtained by various authors and cited in [12]. We compared the experimental BYs with the calculated data [12] and we found reasonable agreement with $T_r(E)$ of Goto for E > 500 eV [11]. Below 500 eV, the Goto's $T_r(E)$ strongly decreases with energy much more than expected from the BY data. Such effect was also described by Seah [25] on a CMA.

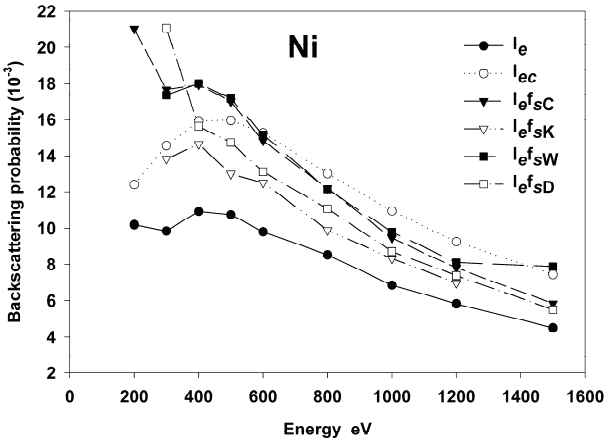

Fig. 2. Comparison of the elastic -backscattering probabilities versus E for Ni. Notations: I_e uncorrected (deduced from Goto's database), $I_e f_{sC}$ SEP corrected with Chen, $I_e f_{sD}$ SEP with Ding, $I_e f_{sK}$ with Kwei, and $I_e f_{sW}$ with Werner. I_{ec} calculated by EPESWIN

Uncorrected experimental IMFPs were deduced from the $i_e(E)$ elastic current, like those presented by Tanuma et al [15], by applying Eqs. (1) and (7). The experimental I_e data have been corrected with the factors f_s, applying f_{sC} (Chen, modified), f_{sK} (Kwei), f_{sD} (Ding) and f_{sW} (Werner) correction factors. The uncorrected λ_u and the

Table 2. SEP correction of uncorrected experimental I_e and SEP- corrected $I_e f_s$ data comparing the fas values of Chen (modified) [17], Kwei [18] Ding [19] and Werner [20]. The following data are presented: the mean deviations ΔI_e between I_e (uncorrected) and I_{ec}, the energy ranges, the mean deviations $\Delta I_e f_s$ between corrected $I_e f_s$ experimental data and I_{ec} for Chen, Kwei, Ding and Werner.

Element	ΔI_e (%)	E range (keV)	$\Delta I_e f_{sC}$ (%)	$\Delta I_e f_{sK}$ (%)	$\Delta I_e f_{sD}$(%)	$\Delta I_e f_{sW}$(%)
Si	24.15	0.4 – 1.2	5.45	12.22	-	4.53
Ni	34.4	0.4 – 1.2	7.12	15.73	9.58	9.58
Cu	31.8	0.4 – 1.5	10.98	14.14	12.03	-
Ag	30.8	0.4 – 1.2	9.28	14.8	9.22	14.5
Au	20.6	0.4 – 1.2	10.53	13.16	12.1	-

corrected λ_{eco} IMFP data were deduced from I_e and from the SEP- corrected backscattering probabilities. They refer to the angular conditions of the CMA of Goto [9]. The corrected IMFPs were determined from the $I_e f_s$ corrected data, applying the EPESWIN software. The λ_u uncorrected, λ_i calculated from (TPP-2M) [7] and the corrected λ_{ecoC} (Chen), λ_{ecoD} (Ding), λ_{ecoK} (Kwei) and λ_{ecoW} (Werner) IMFP results are presented in Figs. 3 to 7 for Si, Ni, Cu, Ag, and Au.

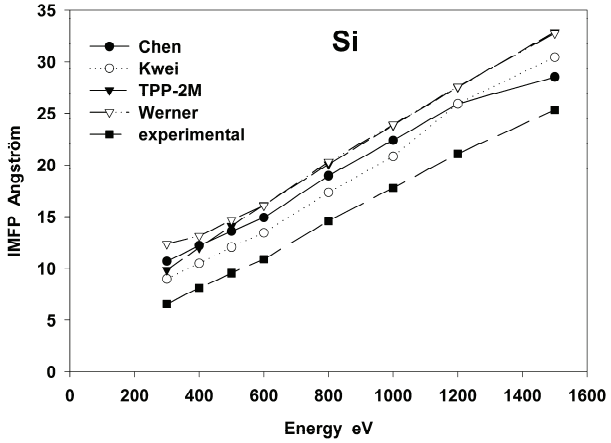

Fig.3. Comparison of IMFPs for Si. Notations of the IMFP curves: experimental (uncorrected), SEP corrected IMFP plots for Chen, Kwei, Ding, and Werner. Calculated IMFPs from the TPP-2M formula. The curves TPP-2M and Werner are overlapping.

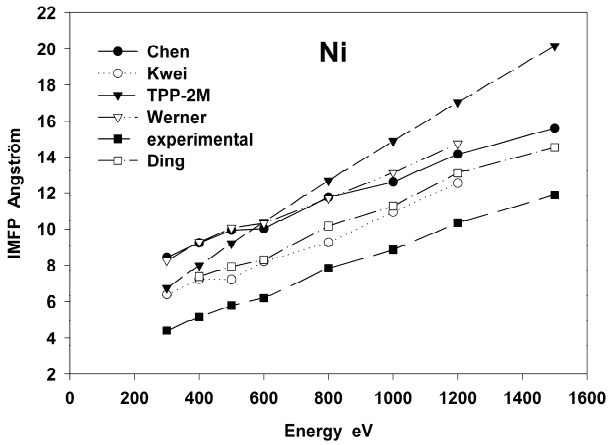

Fig. 4. Comparison of IMFPs for Ni. Notations of the IMFP curves: experimental (uncorrected), SEP corrected IMFP plots for Chen, Kwei, and Werner. Calculated IMFPs from the TPP-2M formula.

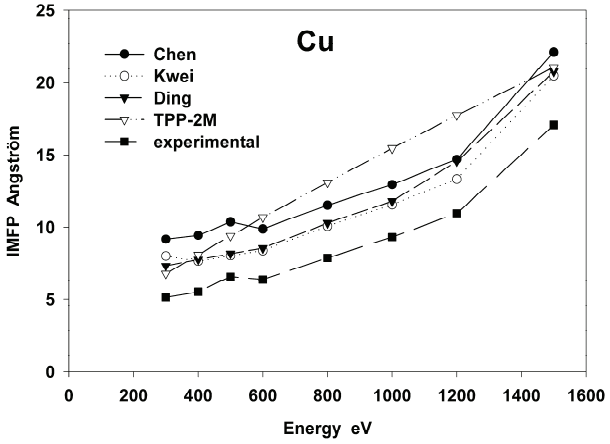

Fig. 5. Comparison of IMFPs for Cu. Notations of the IMFP curves: experimental (uncorrected), SEP corrected IMFP plots for Chen, Kwei, and Ding. Calculated IMFPs from the TPP-2M formula.

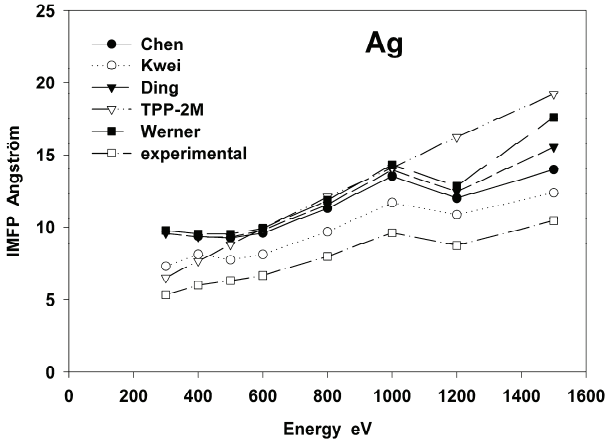

Fig. 6. Comparison of IMFPs for Ag. Notations of the IMFP curves: experimental (uncorrected), SEP corrected IMFP plots for Chen, Kwei, Ding, and Werner. Calculated IMFPs from the TPP-2M formula.

The following results were found: The uncorrected λ_u IMFPs < λ_i calculated from the TPP-2M formula [7]. The mean deviations $\Delta\lambda_u$ between λ_u and λ_i are calculated with Eq. (3). They are 20-34%. The SEP- corrected IMFP λ_{eco} data resulted in better agreement with the λ_i TPP-2M data. The mean deviations $\Delta\lambda_{eco}$ from the λ_i data for the five elements and for the f_s correction factors of Chen (modified), Kwei, Ding, and Werner are calculated with Eq. (3). Results are summarized in the Table 3.

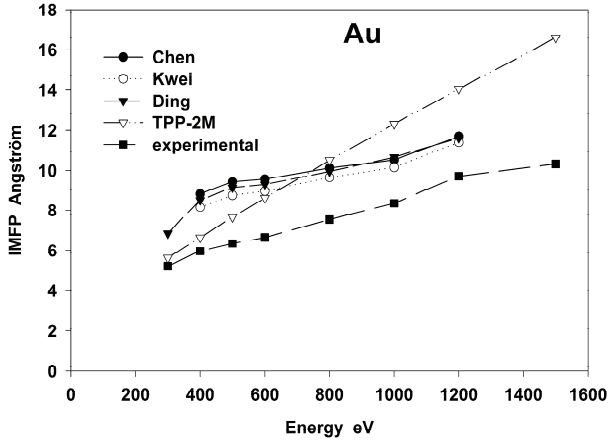

Fig. 7. Comparison of IMFPs for Au. Notations of the IMFP curves: experimental (uncorrected), SEP corrected IMFP plots for Chen, Kwei, and Ding. Calculated IMFPs from the TPP-2M formula.

4. Discussion and conclusions

We emphasize, that the results presented in the Figs. 3-7 and in Tables 2 and 3 refer to the angular conditions of Goto's CMA [9]. The elastic peak I_{ec} and the IMFPS were calculated applying the EPESWIN software [4]. For all five elements, the SEP correction resulted in considerable improvements of the derived IMFPs, SEP-corrected elastic- peak intensities, and IMFPs. Comparing the mean deviations between the uncorrected elastic peaks and IMFPs, the SEP corrected $I_e f_s$ and IMFPs with the calculated I_{ec} and λ_i calculated data, the efficiency of the SEP correction was estimated, using Eq. (3). The efficiency values are the $\Delta\lambda_{eco}$ data in Table 3. This resulted in 3.7% for Si (Werner). For Ni 6% (Nagatomi, Werner), for Ag 8.2% (Chen modified), for Cu, and Au 11.5-12.5% was found for the material parameters of Chen (modified), Kwei, Ding and Werner. Very good SEP- correction on Si was achieved for the energy range E= 300-1200 eV. Quite efficient was the SEP- correction for Ni, E = 300-800 eV.

For Ag and Au, the elastic -peak can exhibit considerable changes of intensity within the angular range α_e=42.3±6° of the CMA. As shown in the Figs. 3-7, fluctuations can be observed of the corrected IMFPs in different energy ranges. This might be a reason of less satisfactory SEP- corrections. The SEP correction was made by averaging over the experimental energy range (0.4-1.2 keV), and applying Eq. (5) and Eq. (6).

Testing of the material parameters of Kwei, Chen (modified), Ding and Werner could be improved by AREPES (angular) experiments [1,22]. Hereupon a new problem of the SEP material parameters should be faced. Our experimental procedure is based on the database of Goto [10]. The same procedure was used by Nagatomi [24] and Tanuma *et al.* [15]. The database refers to the surface conditions (composition) of the samples. They can be affected by contamination from the residual atmosphere even for the 10^{-10} mbar pressure range during the experiments. The ideal SEP parameters refer to atomically clean and flat surfaces. The roughness of the surface can be affected by ion-bombardment cleaning [1].

5. Note added in proof

After submission of our manuscript, Nagatomi sent us his recent paper [26]. Nagatomi and Goto succeeded in the perfect solution; they determined the IMFP and the SEP parameter for Ni. Their IMFPs are in good agreement with the TPP-2M data. Their material parameter a_{chNi} = 4.3 was confirmed. Multiple surface excitation losses occur. The anomaly for Ni in our Figs. 2 and 4 can be explained with the Fig.7 of Nagatomi.

Table 3. Comparison of the deviations $\Delta\lambda_u$ (uncorrected), $\Delta\lambda_{eco}$ SEP corrected and the λ_i TPP-2M IMFPs, applying correction factors using material parameters of Chen (λ_{ecoC}), Kwei (λ_{ecoK}), Ding (λ_{ecoD}) and Werner (λ_{ecoW}).

Element	$\Delta\lambda_u$ %	$\Delta\lambda_{ecoC}$ %	$\Delta\lambda_{ecoK}$ %	$\Delta\lambda_{ecoD}$ %	λ_{ecoW} %
Si	24.7	5.59	11.25	-	3.71
Ni	30.4	6.3	12.76	9.44	5.84
Cu	32.95	11.48	12.92	13.78	-
Ag	22.4	8.22	35.5	19.1	10.66
Au	22.4	13.7	12.6	12.5	-

6. Summarizing our results

SEP correction of EPES experiments achieved considerable approach of the uncorrected elastic-backscattering probabilities to calculated I_{ec} data for Si, Ni, Cu, Ag and Au over the 400-1200 eV energy range. The same SEP-correction improved agreement between IMFPs from the measured EPES data with values from the TPP-2M formula. The best corrections were achieved with the modified material parameters of Chen [17], Nagatomi [24] and of Werner [20]. They are characterized by the mean deviation between the SEP corrected λ_{eco} and λ_I from TPP-2M. The mean deviations were found to be 3.7% for Si, nearly 6% for Ni, nearly 8% for Ag ands about 12% for Cu and Au. In general the mean deviations are not considerably different when using the material parameters of Chen (modified), Ding and Werner. Our work was confined to the CMA of Goto [9].

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