Journal of Surface Analysis Vol.13 No. 2 (2006) pp. 200 - 205
T. Iyasu et al. Construction of a data base for secondary electron emission by a novel approach based on ...

Construction of a data base for secondary electron emission by a novel approach based on Monte Carlo simulations

T. Iyasu and R. Shimizu*
Department of Information Science, Osaka Institute of Technology
1-79-1, Kitayama, Hirakata-shi, Osaka 573-0196, Japan
*shimizu@is.oit.ac.jp

(Received: April 8, 2006 ; Accepted: September 5, 2006)

A novel approach is proposed for a construction of data base for the secondary electron emission which is described by the equation

$$\delta(E_P) = k\int_0^{\infty}\left[\frac{dE}{dz}\right]_{E_P} \exp(-\alpha z)\, dz \ .$$

where k and α are the secondary emission coefficient and absorption coefficient, respectively, to be derived for the data base. E_P is the primary energy of incident electrons and $[dE/dz]_{E_P}$ the energy dissipation with respect to depth which is to be obtained from Monte Carlo calculation.

This approach is based on the use of theoretical secondary electron yield, $\delta(E_P)$, to derive and by comparing with experimental secondary electron yield, $\delta_{\exp}(E_P)$. Usually, $\delta(E_P)$ is characterized by the two physical quantities, the maximum of secondary yield, δ_m, and the primary energy E_m which provides δ_m, i.e., $\delta_m = \delta(E_m)$. These physical quantities, k, α, δ_m, and E_m have been believed to be independent of one another.

The present study, however, has revealed that α and E_m are closely correlated, enabling us to derive and by comparing $\delta(E_P)$ with either experimental $\delta_{\exp}(E)$ or a set of experimental values of δ_m and E_m. A preliminary construction of a data base of (k, α) for 25 materials is presented.

1. Introduction

The secondary electron yield, δ, the average number of secondary electrons per number of primary electrons, is described by a simple equation [1,2],

$$\delta(E_P) = k\int_0^{\infty}\left[\frac{dE}{dz}\right]_{E_P} \exp(-\alpha z)\, dz \qquad . \qquad (1)$$

where $[dE/dz]_{E_P}$ is the energy dissipation with respect to depth of an incident electron of primary energy E_P. α is the absorption coefficient which was used by Bruining in the theory of secondary electron emission [2] and has been widely used in studies of secondary electron emission since then [3], to describe the probability of secondary electrons generated at the depth, z, to emerge from the surface. k is the secondary emission coefficient, that describes the efficiency with which energy dissipated in a layer, dz, at the depth z is converted to generation of secondary electrons. So, k and α may be regarded as such material constants such as the Richardson constant and the work function in thermionic emission [4]. Precise knowledge of the set (k, α), therefore, allows a Monte Carlo simulation approach to describe the secondary electron emission with considerable quantitative accuracy for various boundary conditions of practical importance for scanning electron microscopy (SEM), in particular, critical dimension (CD)-SEM [5].

The question may arise as to why the absorption coefficient, α, has been used here even though the inelastic mean free path, λ_{inel}, has been widely used in surface electron spectroscopies [6], e.g., Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS) etc. The reason is that a secondary electron undergoing inelastic scattering processes still has the possibility to remain as a secondary electron because the energy spectrum of the secondary electrons spreads over the energy region from 0 to ~50 eV [7]. Furthermore, those secondary electrons with higher energies often cause a collision cascade of secondary electrons leading to the generation of other secondary electrons with lower energies. The concept of an absorption coefficient or its inverse, α^{-1}, is, therefore, not as simple as the case of the inelastic mean free path which describes the attenuation of signal electrons, where the decay of signal intensity is simply described by $\exp(-s/\lambda_{inel})$ with s being the path length.

Despite the fundamental importance of understanding secondary electron emission, only very few experimental

data for α have been reported so far, probably due to the laborious task to obtain the absorption coefficient in experiment. Seiler and Staerk [8] measured the decrease of secondary electron intensity by irradiating a Cu-specimen surface with an electron beam, which resulted in the growth of a hydrocarbon film on the specimen with increasing irradiation time. They estimated the inverse of absorption coefficient for the hydrocarbon as α^{-1} (hydrocarbon) $\cong$ 10 nm from the decay curve of the secondary electron intensity plotted as a function of the hydrocarbon film thickness. Bronshtein and Segal [9] have also reported a systematic investigation of α by plotting δ as the ordinate and the backscattering coefficient, η, as the abscissa, a so called $\delta - \eta$ curve, by depositing one material onto a substrate of another material.

Since the experiments to obtain the absorption coefficient are laborious, extensive work can hardly be expected even though up-to-date data on α has been attracting reviewed attention from industry, in particular, plasma display panel (PDP) manufactures. In contrast to α, experimental measurements of $\delta_{\exp}(E_P)$ have been reported continuously [10]. Hence, if the absorption coefficient can be derived from $\delta_{\exp}(E_P)$, the absorption coefficients for materials of practical interest can be determined more easily.

In this paper, we propose a novel approach to derive the absorption coefficient, α, and then, the secondary emission coefficient, k, from comparisons of with theoretical $\delta(E_P)$ values obtained from Monte Carlo calculations. The secondary emission yield is characterized by the two physical quantities, the maximum secondary yield δ_m, and corresponding primary energy E_m [10]. Verification of the approach and its application to 25 materials as a preliminary construction of a data base of (α, k) are presented. It is also noted that a similar attempt to describe the secondary electron yield as a function of E_P and atomic number Z for 44 elements has been reported by Lin and Joy [11].

2. Monte Carlo calculation

First, a Monte Carlo calculation was performed to obtain $[dE/dz]_{E_P}$. The model is based on the use of screened Rutherford formula and Bethe's stopping power equation as follows [12]:

The elastic scattering process is described by

$$\frac{d\sigma}{d\Omega} = \frac{1}{4}\left(\frac{Ze^2}{mv^2}\right)^2 \cdot \frac{1}{\left(\sin^4\frac{\omega}{2} + y_0^2\right)} \quad ,$$
$$y_0 = \frac{\hbar\lambda_0}{2mv} \quad , \quad \lambda_0 = \frac{Z^{1/3}}{0.885a_0} \quad . \qquad (2)$$

m, v and ω are mass, velocity and polar scattering angle of an incident electron, respectively, a_0 is the Bohr radius and Z the atomic number of the target atom.

Bethe's equation in the continuous slowing down approximation with Joy's correction factor [12] to describe inelastic scattering is

$$\frac{dE}{dz} = -\frac{2\pi e^4}{E}\cdot\frac{\rho N_A Z}{A}\cdot\ln\frac{1.166E}{J^*} \quad , \quad J^* = \frac{J}{1+0.85\frac{J}{E}}$$
$$J = 9.76Z + 58.8Z^{-0.19} \quad for \quad Z \geq 13$$
$$J = 11.5Z \quad for \quad Z \leq 12 \quad . \qquad (3)$$

where the mean ionization energy J in the original Bethe's equation was replaced by J^* [13] which allows us to extend the equation to the energy range below 1 keV. Z, A, ρ and N_A are the atomic number, atomic weight, density and Avogadro number, respectively.

Then, $[dE/dz]_{E_P}$ was calculated for different primary energies under the boundary condition corresponding to the experiment as seen in Fig. 1. $\delta(E_P)$ is, then, calculated for different values of α to find the best fit value of the absorption coefficient, α_0, by comparing with the relevant experimental $\delta_{\exp}(E_P)$.

Note, the comparison has revealed that these exists a very close correlation between α_0 and E_m. This suggests the possibility that one can find a best fit value, α_0, which provides $[E_m]_{MC}$, agreeing well with the experimental $[E_m]_{\exp}$. It has long been believed that E_m and α are independent each other. Actually it has turned out that they are closely correlated to each other.

Once the best fit values of absorption coefficient, α_0, is obtained, one can, then, find a $k-$ value, k_0, which satisfies

$$\delta_m = k_0 \int_0^\infty \left[\frac{dE}{dz}\right]_{E_m} \exp(-\alpha_0 z)\, dz \quad . \qquad (4)$$

The values of k_0 and α_0^{-1} thus obtained are listed in Table 1 and compared with the experimental results, $\alpha_{\exp}^{-1}$, published so far [9].

3. Results and Discussion

3.1 Universal curve plot

Baroody [14] proposed the universal plot, $\delta(E_P)/\delta_m$ versus E_P/E_m curve for various E_P, where he presumed

$$\delta(E_P) = k\int_0^\infty \left[\frac{dE}{dz}\right]_{E_P} \exp(-\alpha z)\, dz \quad . \qquad (5)$$

δ_m is the maximum secondary yield to be obtained at primary energy $E_P = E_m$, i.e. $\delta_m = \delta(E_m)$.

Among the physical quantities k, α, δ_m and E_m, the present work has revealed that there exists a very close correlation between α and E_m which have long been believed to be independent of each other. This, therefore, enables α to be obtained from E_m by comparing theoretical

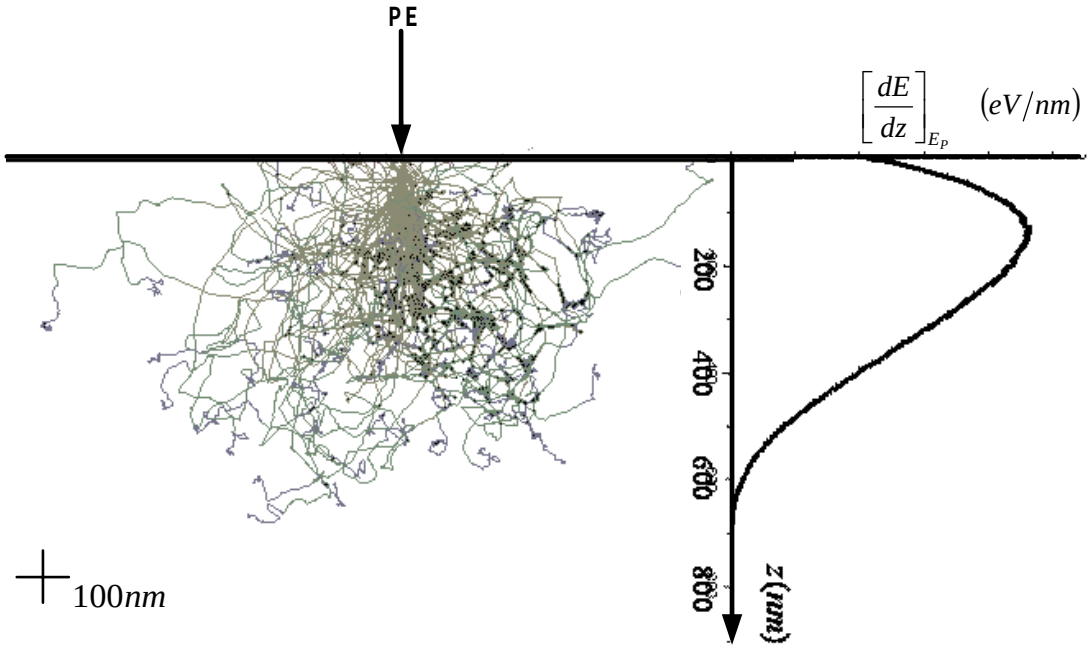

Fig.1 Trajectories of incident electrons of 1keV in a Cu target. The energy dissipation in-depth is also depicted.

Table1 Comparison of theoretical absorption coefficients, α_0, with experimental values, α_{ex}[9].

Material	α_0^{-1} (nm)	α_{ex}^{-1} (nm)	k_0
Be	1.1	0.7	0.017
Ag	1.4	1	0.01
Pt	0.4	0.7	0.086
Bi	0.8	0.8	0.027

$\delta(E_P)$ values with experimental $\delta_{\exp}(E_P)$ measurements or more directly with the experimental E_m.

According to the experimental universal curve plot proposed by Baroody [14], theoretical universal curves were obtained from Monte Carlo calculations for Be, Ag, Pt and Bi. Results are depicted in Fig. 2. All of the δ/δ_m versus E_P/E_m curves for each element plotted for different values of α overlap each other, forming almost a single curve. In other words, all the curve plotted for different α values depicted for each element in Fig. 2 are represented by a single curve in Fig. 3. This strongly indicates that α is very closely correlated with E_m, suggesting that α can be derived from E_m by using $[dE/dz]_{E_P}$ which is obtained from Monte Carlo calculations.

The difference in the shapes of the curves for the four elements in Fig. 3 can be attributed to the Rutherford scattering formula not being a good approximation to describe elastic scattering for heavier elements in the low energy region below 1 keV [15]. A systematic M.C. calculation based on the use of Mott scattering cross-sections has provided better agreement among the curves for different elements including the above four elements, which are then represented by a single universal curve more reasonably [16].

Consequently the results shown in Fig. 2 indicate that the present approach describes $\delta_{\exp}(E_P)$ sufficiently for the derivation of α_0 and k_0 from the database of δ_m and E_m which has been published [17] so far for a number of materials of practical use.

3.2 Preliminary construction of data base for (k,α)

Since a set of δ_m and E_m has been reported [17] for number of materials, we have applied the present approach for these materials to derive k_0 and α_0^{-1}. The results are listed in Table 2, in which up-to-date data for TiO_2 are also presented. It is found that α_0^{-1} values are around 1nm or less except for a few metals which are liable to being oxidized under the experimental conditions of insufficient high vacuum. In fact, this was the situation when these measurements were performed. Hence, α_0^{-1} values of these metals are probably not so accurate as those of the other materials.

The α_0^{-1} values in Table 1 may, therefore, lead to the common feature that the α_0^{-1} values are around 1nm or less, with most between ~1 and ~0.5 nm though this finding calls

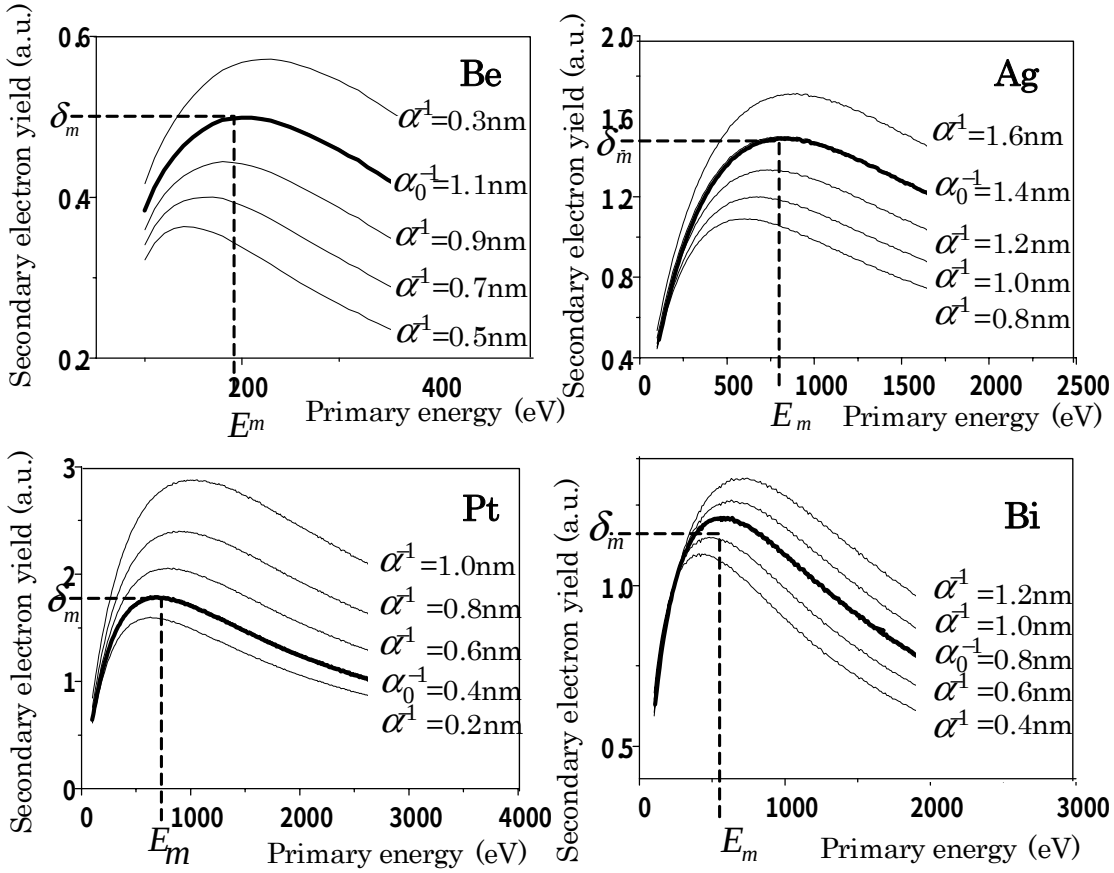

Fig.2 Theoretical $\delta(E_P)$ obtained from Eq.4 for different values of absorption coefficient: (a)Be, (b)Ag, (c)Pt and (d)Bi.

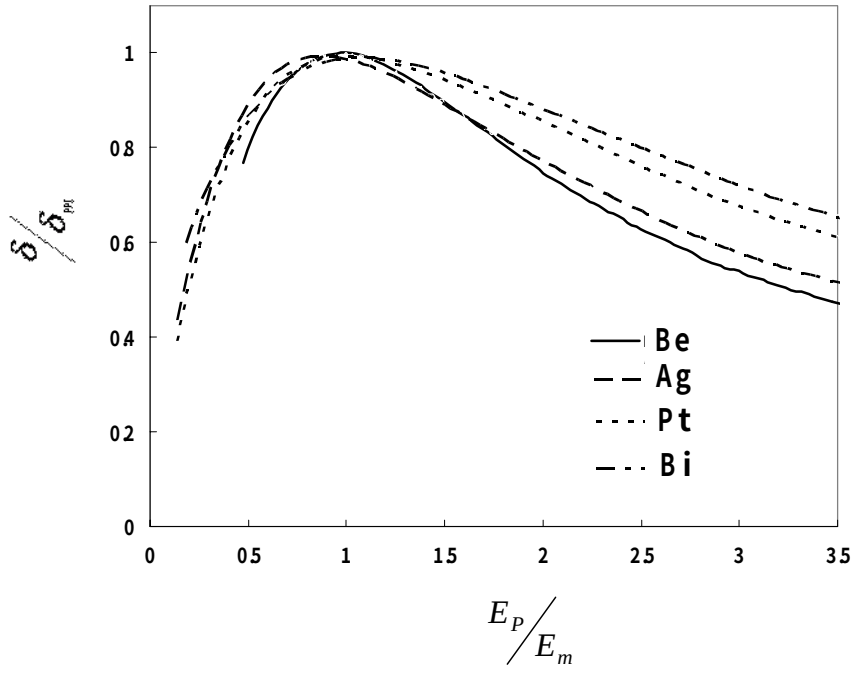

Fig.3 Theoretical universal curve plot as a function of absorption coefficient for Be, Ag, Pt and Bi

for more systematic investigation to obtain more reliable data for E_m and δ_m with modern surface analytical instruments before construction of the data base of k_0 and α_0. Since measurements of δ_m and E_m with modern surface analytical instruments should be easier than in the early days of secondary electron emission studies, the construction of a data base of k and α is now becoming attainable.

As an example of this proposal, experimental results for Ti and TiO_2 have recently been obtained by the authors [17] under the same experimental conditions. The results clearly indicate that the α_0^{-1} value for TiO_2 is larger than that for Ti. This supports the idea [3] that the escape depths of the secondary electrons in insulators and semiconductors are longer than those for metals because the interaction with conduction electrons is much smaller.

4. Summary

The present paper reports a basic study of the secondary electron yield described by

$$\delta(E_P) = k\int_0^{\infty}\left[\frac{dE}{dz}\right]_{E_P} \exp(-\alpha z)\, dz \quad . \qquad (6)$$

We proposed a novel approach to derive two physical quantities of practical importance, i.e. the secondary electron absorption coefficient α and the secondary emission coefficient k. The results are summarized as follows:

(1) A study of Eq. 5 led to the finding that an intrinsic correlation does exist between α and E_m.
(2) Based on this finding, we proposed a novel approach to derive the absorption coefficient α and the secondary emission coefficient k from a database of δ_m and E_m by using the energy dissipation with depth $[dE/dz]_{E_P}$ which is obtained from Monte Carlo calculations.
(3) Once a data set (α_0 and k_0) for a specific material of practical interest is provided by the present approach, one can extend the quantitative calculation of secondary electron emission under different experimental conditions of practical use, e.g. different primary energies E_P and angles of incidence θ of primary electron beam by using

$$\delta(E_P,\theta) = k_0\int_0^{\infty}\left[\frac{dE}{dz}\right]_{E_P,\theta} \exp(-\alpha_0 z)\, dz \quad . \qquad (7)$$

where $[dE/dz]_{E_P,\theta}$ can easily be obtained from Monte Carlo calculation with sufficient accuracy. An application of Eq. (6) for critical dimension (CD) SEM has been under examination to find the best choice of E_P and θ for eliminating the charging up effect, and the result will be published shortly.

Table2. Theoretical absorption coefficient and k values obtained from Eq.4 after δ_m and E_m are determined by comparison with experimental data from references (10) and (17).

	Inverse of Absorption coefficient α_0^{-1}(nm)	k	Experiment[10]	
			δ_m	E_m (eV)
Be	1.1	0.017	0.5	200
Mg	1.9	0.012	0.95	300
Al	1.3	0.014	1	300
Si	1.0	0.024	1.1	250
Ti	0.5	0.046	0.9	280
Fe	0.5	0.048	1.3	400
Co	1.1	0.011	1.2	600
Ni	0.8	0.017	1.3	550
Cu	1.1	0.011	1.3	600
Ga	1.2	0.017	1.55	500
Ge	1.2	0.013	1.15	500
Zr	0.4	0.093	1.1	350
Ag	1.4	0.010	1.5	800
Cd	0.5	0.047	1.1	450
Sn	0.9	0.024	1.35	500
Sb	1.4	0.014	1.3	600
Ba	1.4	0.015	0.8	400
W	0.4	0.070	1.4	650
Pt	0.4	0.086	1.8	700
Au	0.7	0.022	1.4	800
Hg	0.6	0.036	1.3	600
Pb	0.5	0.048	1.1	500
Bi	0.8	0.027	1.2	550
Th	1.3	0.012	1.1	800

	δ_m	E_m (eV)	α_0^{-1} (nm)	k
TiO_2	1.2	260	0.7	0.028

From reference (17)

Acknowledgements

One of the authors(R.S) gratefully acknowledges enjoying the cooperation of Dr. Yoshihara for international standardization over 25 years, starting from joining the VAMAS-TWA-Surface Chemical Analysis in 1985 and, then, establishment of ISO-TC/201(Surface Chemical Analysis) in 1992, in which Dr. Yoshihara has been leader of the Japanese delegation and later Chairman.

The authors wish from the bottom of their hearts that Dr. Yoshihara will enjoy continuous success in his new career in industry.

References

[1] O. Hachengerg and W. Brauer, *Adv . Electronics and Electron Physics,* **11**, 413 (1959).
[2] H. Bruining, *Physics and Application of Secondary Electron Emission,* Pergamon Press, London, 1954.
[3] A. J. Dekker, in *Solid State Physics* vol. 6, F. Seitz and D. Turnbull Eds., Academic Press, New York, 1959, p.251.
[4] A. J. Dekker, *Solid State Physics*, Prentice Hall Inc. 1957, chap.17.
[5] U. Dersch, A. Korn, C. Engelmann, C. G. Frase, W. Haessler-Grohne, H. Bosse, F. Letzkus, J. Butschke, Metrology, Inspection, and Process Control for Microlithography XIX. ed. R. M. Silver, SPIE, Bellingham. WA, 2005, p.632.
[6] S. Tanuma, C. J. Powell and D. R. Penn, *Surf. Interface Anal.*, **17**, 911 (1991).
[7] H. Seiler, *Z. Angew. Phys.* **22**, 249 (1967).
[8] H. Seiler and M. Staerk, *Z. Phys*. **183**, 527 (1965).
[9] I. M. Bronshtein and R. B. Segal, *Sov. Phys. Doklady*, **3**,1184(1958).
[10] *CRC Handbook of Chemistry and Physics*, CRC Press, Boca Raton, Florida, published every two years).
[11] Y. Lin and D. Joy, *Surf. Interface Anal.*, **37**, 895 (2005).
[12] '*Use of Monte Carlo Calculations in Electron Probe Microanalysis and Scanning Electron Microscopy'* , Eds. K. F. J. Heinrich, D. E. Newbury and H. Yakowitz, NBS Special Publication 460, NBS. Gaithersburg, 1975.
[13] D. C. Joy and S. Luo, *Scanning*, **11**,176 (1989).
[14] E. M. Baroody, *Phys. Rev*. **78**, 780 (1950).
[15] A. Jablonski, F. Salvat and C. J. Powell, *J. Phys. Chem. Ref. Data*, **33,** 409 (2004).
[16] Z. J. Ding and X. D. Tang, *J. Appl. Phys.*, **89**, 718 (2001).
[17] T. Iyasu, M. Inoue, H. Yoshikawa and R. Shimizu, *Surf. Interface Anal.* **38** (2006), in press.