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# Estimation of Inelastic Mean Free Paths in Au and Cu from Their Elastic Peak Intensity Ratios without IMFP Values of Reference Material in The 200 – 5000 eV Energy Range

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We have determined electron inelastic mean free paths (IMFPs) and surface-electronic excitation parameters (SEPs) of Au and Cu in the 200 – 5000 eV from their elastic peak intensity ratios without reference IMFP values. This proposed method does not require the IMFP values of the reference material. The measurements of elastic peak intensities of these elements were done with noble CMA system. The elastic peak intensities were also calculated from two parameter sets (IMFPs and SEPs for both elements) with Monte Carlo method. By comparison of these two EPI ratios as changing the parameter sets, we have estimated their IMFPs and SEPs in the 200 – 5000 eV energy range. The resulting IMFPs of Au and Cu were in good agreement with optical IMFPs over 1000 eV energy range.

The elastic and inelastic scatterings of electron in solid are very important for surface electron spectroscopies. Especially, it is essential to describe the attenuation rate of the electron signal due to inelastic scattering events in a solid. Therefore, the knowledge of the electron inelastic mean free paths (IMFPs) for low-energy electrons in solids is required for quantitative surface analysis by AES and XPS as well as for determining the surface sensitivity of other electron-spectroscopic methods of surface characterization.

In a series of the papers Tanuma *et al.* calculated IMFPs for 50 - 2000 eV electrons in a group of 41elemental solids [1-3], a group of 15 inorganic compounds [4], and a group of 14 organic compounds [5] from their optical energy loss functions with Penn algorithm [7]. They fitted the calculated IMFPs for the groups of elements and organic compounds to a modified form of the Bethe equation for inelastic electron scattering in matter and found that the four parameters in this equation could be related empirically to several material parameters [5]. The resulting general formula TPP-2M for IMFPs in solids could be used to estimate IMFPs for other materials in the 50 – 30,000 eV energy range.

The evaluation of the accuracy of theoretical IMFPs for a given material is very difficult because it depends on the accuracy of the particular energy loss function (ELF) on which the calculation is based and of the approximations used in Penn algorithm. It is valuable to compare calculated IMFPs and those obtained from experimental method in order to assess the consistency of the IMFP values.

Elastic peak electron spectroscopy (EPES) is a useful method for experimental determination of IMFPs [6]. In most EPES measurements it has been convenient to compare elastic peak intensities from a target material with those of a suitable reference material. With this method the instrumental factor such as mesh transmission efficiency could be removed. However, it requires the IMFP values of the used reference material. Alternatively, the resulting IMFPs depend on the IMFPs of reference material. On the other hand, IMFPs can be directly determined from absolute measurement of elas-

tic-backscattering coefficient of incident electrons. With this approach, it is necessary to make several corrections such as surface-electronic excitation, transmission efficiency etc. Generally it is very difficult to measure the transmission efficiency of the analyzer with high accuracy. Then, we have developed a new method to determine IMFPs of the target material from elastic peak intensity ratios without IMFPs of the reference.

The energy dependence of elastically backscattered primary electrons were measured for Au and Cu with a novel cylindrical mirror analyzer (CMA) [8,9] in the 50 – 50 000 eV energy range. The details of the measurement were already published [10]. Figure 1 shows the measured peak intensity ratios of Au to Cu as a function of primary energy.

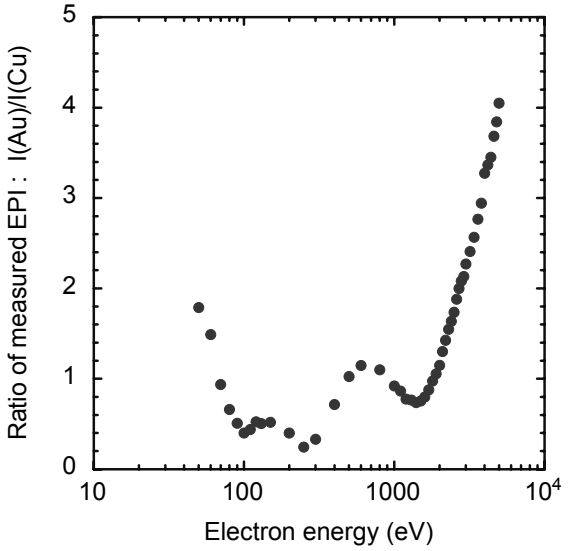


Fig. 1. Ratios of measured elastic peak intensities for Au to Cu as a function of primary electron energy.

The intensity ratio of elastically backscattered primary electrons for Au and Cu, $I^{Au}/I^{Cu}$, can be calculated from the following equation:

$$\left(\frac{I^{Au}}{I^{Cu}}\right) = \frac{f_S^{Au}}{f_s^{Cu}} \frac{\int_0^\infty (d\eta/dS)^{Au} / N_0^{Au} \exp(-S/\lambda_{Au}) dS}{\int_0^\infty (d\eta/dS)^{Cu} / N_0^{Cu} \exp(-S/\lambda_{Cu}) dS} = \frac{f_S^{Au}}{f_s^{Cu}} \frac{H(E,\lambda_{Au})}{H(E,\lambda_{Cu})} \quad (1)$$

where $f_s$ is a surface-electronic excitation (SEE) correction factor, $d\eta/dS$ is the path-length distribution of elastically backscattered electrons, $N_0$ is the number of input electrons, $S$ is total path length, and $\lambda$ is the electron inelastic mean free path. The $H(E,\lambda)$ is the reflection coefficient of the incident electron at primary energy $E$ and IMFP $\lambda$. The $d\eta/dS$ were calculated with Monte Carlo (MC) method. In this calculation, we require the differential elastic-scattering cross sections for their elements and energies at elastic peak intensities measured. The differential cross section $d\sigma(E)/dq$ for elastic scattering used in the calculations were cited from the database of NIST [11]. In the calculation of $d\eta/dS$, MC calculation traces the input electrons until they are scattered back from the target specimen or until their total path length becomes larger than $10\times\lambda_{TPP\text{-}2M}$, where $\lambda_{TPP\text{-}2M}$ denoted the IMFP values calculated from TPP-2M [5]. We collect the backscattered electron from the target within 42.3±6° to get the histogram $d\eta/dS$ until at least over 10 000 electrons are detected.

The dependence of reflection coefficient $H(E,\lambda)$ on IMFPs for Au and Cu were calculated in the 50 – 5000 eV energy range. The calculated results at 200, 1000, and 5000 eV were shown in Fig. 2. From this figure, we see that the $H(E,\lambda)$ values are monotonically increased according to the increase of IMFP. Then, we have fitted $H(E,\lambda)$ with quintic equation. The results were already shown in Fig. 2 as solid lines. The root mean square (rms) differences for the fits were within 0.2% in the range of $\lambda$ from $0.2\lambda$ to $3\lambda$. Then, $H(E,\lambda)$ can be expressed by the quintic equation of $\lambda$ at primary energy $E$ with high accuracy.

Over 200 eV, the energy dependence of IMFP can be expressed by the following simple Bethe equation [1]

$$\lambda(E) = \frac{E}{E_p^2 \beta \ln(\gamma E)} \quad (2)$$

where $E_p$ is the bulk plasmon energy (in eV) of the material, and $\beta$ and $\gamma$ are the fitting parameters for the material. Then, the energy dependence the reflection coefficient $H(E,\lambda)$ can be expressed as a function of $\beta$ and $\gamma$.

The surface-electronic excitation correction factor $f_s$ for EPES can be described as

$$f_S = \exp\left[-P_S(\alpha_{in},E)\right]\exp\left[-P_S(\alpha_{out},E)\right] \quad (3)$$

where $P_s$ is the surface-excitation probabilities (SEP) for electrons entering or leaving the solid. $\alpha_{in}$ is the electron incident angle to the surface normal and $\alpha_{out}$ is the angle

of detected electrons. In the present study, 0 and 42.3 degrees were used for $\alpha_{in}$ and $\alpha_{in}$, respectively. Werner *et al.* [12] proposed the following formula for estimating the SEP in a material based on the Oswald equation [13].

$$P_S = \frac{1}{a\sqrt{E}\cos(\alpha)+1} \tag{4}$$

where $\alpha$ is the angle of electron incidence or emission with respect to the surface normal and $a$ is a material parameter.

From equations (1) to (4), then, the intensity ratio of elastically backscattered primary electrons for Au and Cu, $I^{Au}/I^{Cu}$, can be expressed by the following equation using three parameters $a$, $\beta$ and $\gamma$ for the material over 200 eV energy region.

$$\left(\frac{I^{Au}}{I^{Cu}}\right) = \frac{f_S^{Au}}{f_s^{Cu}}\frac{H(\lambda_{Au})}{H(\lambda_{Cu})} \approx \frac{\exp\left[-1/\left(a_{Au}\sqrt{E}+1\right)-1/\left(0.74\sqrt{E}a_{Au}+1\right)\right]}{\exp\left[-1/\left(a_{Cu}\sqrt{E}+1\right)-1/\left(0.74\sqrt{E}a_{Cu}+1\right)\right]} \times \frac{H\left\{E/E_{p,Au}^2\beta_{Au}\ln(\gamma_{Au}E)\right\}}{H\left\{E/E_{p,Cu}^2\beta_{Cu}\ln(\gamma_{Cu}E)\right\}} \tag{5}$$

where $H$ can be expressed by the quintic equation of $\lambda$ or $E/E_p{}^2\beta\ln(\gamma E)$, and their coefficients at energy $E$ were determined by the curve fits as stated above.

Then, we optimized the two sets of three parameters ($a$, $\beta$ and $\gamma$ for Au and Cu) in the 200 – 5000 eV energy range so as to $\Sigma\{(I^{Au}/I^{Cu})_{cal}/(I^{Au}/I^{Cu})_{obs}-1\}^2\rightarrow$min with "solver" command in EXCEL. The calculated results were shown in Fig. 3 as a solid line. We see that the calculated values with Eq (5) are in excellent agreement with the measured value. Their rms difference was about 4.3%. The dotted line showed the EPIs without surface-electronic excitation contribution, and these are, of course, smaller than those of measured one. However over 2000 eV, their differences are small.

The resulting IMFPs of Au ($\beta$ = 0.0159, $\gamma$ = 0.456) and Cu ($\beta$ = 0.0111, $\gamma$ = 0.106) were shown in Fig. 4 together with optical IMFPs (solid line) which were calculated

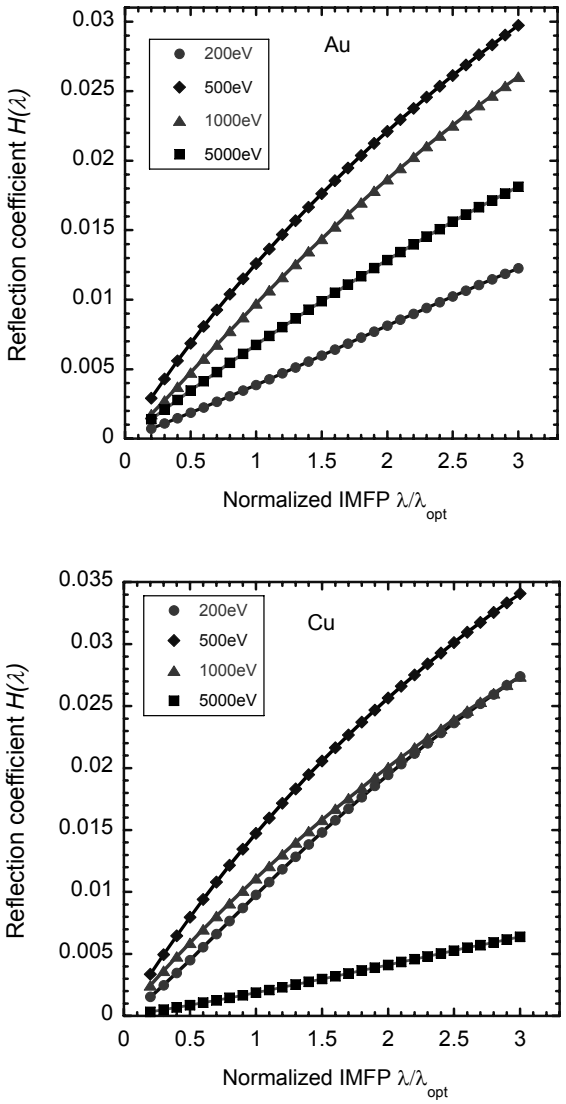


Fig. 2. Dependence of the theoretical reflection coefficient H(l) on the normalized IMFPs for Au and Cu at several energies. Solid marks show the calculated reflection coefficient at several electron energies by MC method. The solid lines show the

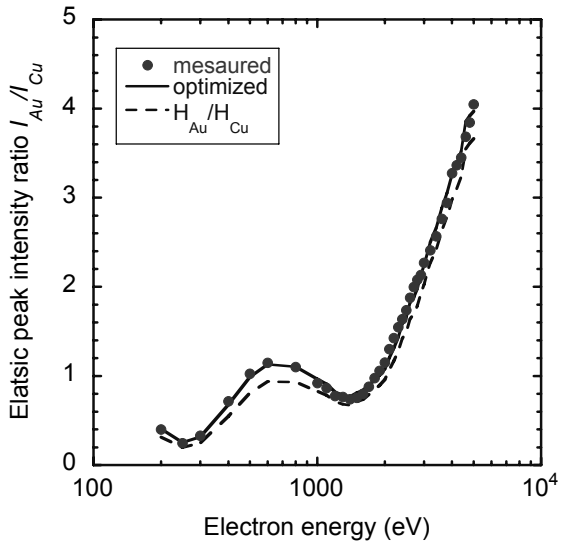


Fig. 3. Calculated elastic peak intensity (EPI) ratios ,$I_{Au}/I_{Cu}$, for Au to Cu with Monte Carlo calculations as a function of primary electron energies. The solid line shows the results of optimization with the proposed method and solid circles show the measured peak intensity ratios. The dotted line indicates the EPIs without surface-electronic excitation correction factor.

from their energy loss function with Penn algorithm. On both elements, the resulting IMFPs were in good agreement with optical IMFPs over 1000 eV. The resulting IMFPs for Cu were smaller than those of optical values especially under 1000 eV. Their rms relative differences between IMFPs in this study and those of optical were 12.5% for Au and 8.4% in the 200 – 5000 eV energy range. These values are slightly larger than the results of IMFP determination with EPES using Ni standard [13].

In this energy region, surface-electronic excitation effect may play important roll. Then, Fig. 5 shows the resulting ratio of surface-electronic excitation factors $f_S^{Au}/f_S^{Cu}$ ($a_{Au}$ = 0.402, $a_{Cu}$ = 0.191) together with that of Werner ($a_{Au}$ = 0.260, $a_{Cu}$ = 0.346) [12]. Form this figure, we see that the SEE on present work was larger than the unity by 20 to 30% under 1000 eV energy range. On the other hand, its effect was smaller than unity over 200 to 5000 eV in Werner results. This is due to the difference of order of the magnitude of surface-electronic excitation effect of Au and Cu on both studies. The details will be discussed elsewhere [14].

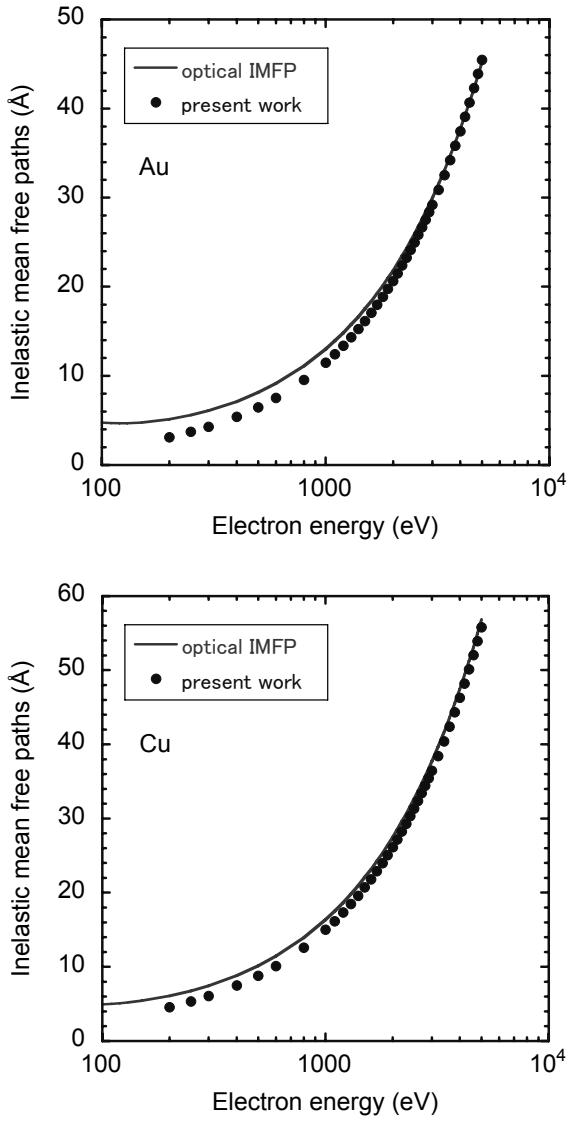


Fig. 4. IMFP values (solid circles) for Au and Cu determined from elastic peak intensity ratios in Fig. 3 as a function of electron energy. The solid line shows IMFPs calculated from Penn

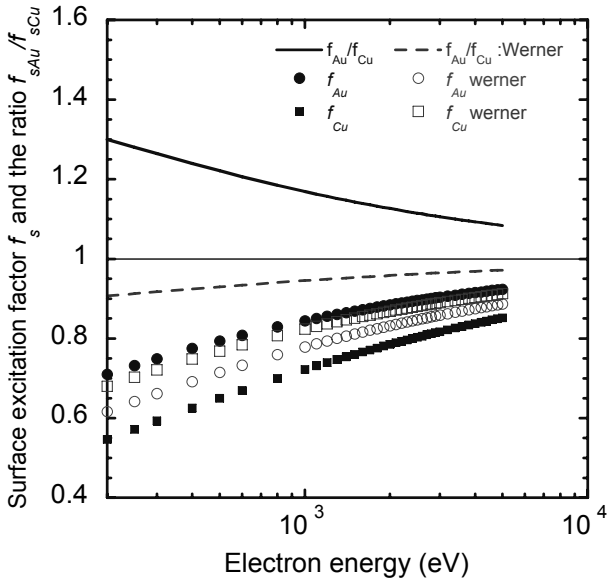


Fig. 5. The energy dependence of the surface-electronic excitation factor $f_s$ for Au and Cu, and their ratios ($f_{Au}/f_{Cu}$). The solid line shows the $f_{Au}/f_{Cu}$ in the present work. The dotted line shows the calculated $f_{Au}/f_{Cu}$ from Werner equation and his $a$ values for Au and Cu. The solid circles and squares represent the $f_{Au}$ and $f_{Cu}$ determined present study. The open circles and squares show the $f_{Au}$ and $f_{Cu}$ calculated from Werner equation and his $a$ values for Au and Cu.

In summary we have determined electron IMFPs of Au and Cu in the 200 – 5000 eV from their elastic peak intensity ratios without reference IMFP values. This used method does not require the IMFP values of the reference material. The resulting IMFPs of Au and Cu were in good agreement with their optical IMFPs(rms differences for Au was 12.5% and that for Cu was 8.4%). This method must be useful to measure the IMFPs of wide variety of materials with EPES in wide energy range.

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