〈연구논문〉

Thickness Measurement of Overlayer Deposited on Single Crystal

Hang-Gi Min and Dae-Hyun Byun

Department of Physics, Hongik University, Seoul 121-791
(Received November 9, 1991)

금속 단결정면에 증착된 층의 두께측정

민항기 · 변대현

홍익대학교 물리학과
(1991년 11월 9일 접수)

Abstract—It is not easy to determine the coverage of deposited overlayers on a single crystal. There are several techniques determining the overlayer thickness. We propose, in this study, a new simple method by using Auger spectra only without any sophisticated thickness monitor. An example of iron overlayers on copper single crystals is also showed.

요 약—표면 연구에서 단결정면 위에 진공 증착된 층의 두께를 측정하는 일은 쉽지 않다. 지금까지 몇 가지 측정방법들이 알려지고 있으나 이들 방법은 섬세한 고가장비를 필요로 한다. 본 연구에서는 단순히 Auger 스펙트럼을 이용하여 두께를 측정할 수 있는 간단한 방법을 제안하고 있다. 또한 구리 단결정면위에 증착된 철 원자의 두께 측정에 관한 예가 제시되고 있다.

1. Introduction

Any piece of condensed matter is necessarily bounded by a surface through which it interacts with the outside world. It is therefore very important for solid-state physicists, chemists, and metallurgists to have a good knowledge of its surface properties. Although a surface is defined by the boundary between any two different states of matter, most activities in surface science are related to the studies of clean solid surfaces and surfaces with a limited amount of additional foreign atoms which are deposited by evaporation or gas reaction.

For the quantitative analysis of surface chemical compositions by Auger electron spectroscopy(AES), it is necesary to determine the coverage of adsorbed overlayers. In order to determine the thickness of an adsorbed material, we need to know the inelastic mean free path(IMFP) of the electrons. The IMFP of the electrons in the crystal is strongly electron energy dependent and to a certain extend meterial dependent too. Also the IMFP is probably dependent on the direction of the crystal because the density of the face and distance between the layers are different.

A lot of work about IMFP has been done statistically using many experimental data by Seah[1] and theoretically by Penn *et al.*[2, 3], Ibach[4] and Powell[5]. We can use the IMFP values from the above references.

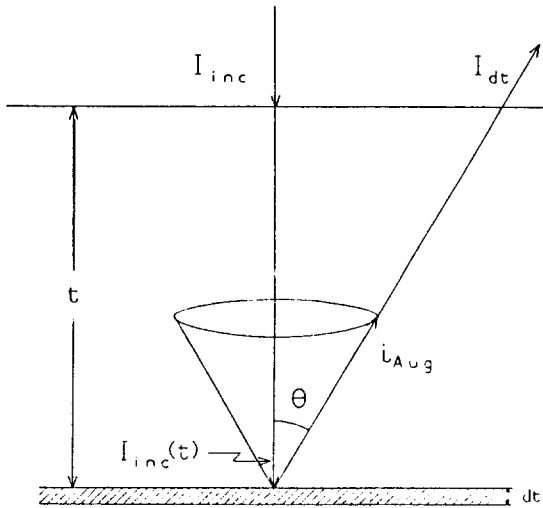## 2. Formulation

The emission current of Auger electrons from

Fig. 1. A schematic representation of clean sample showing quantities used in the text.
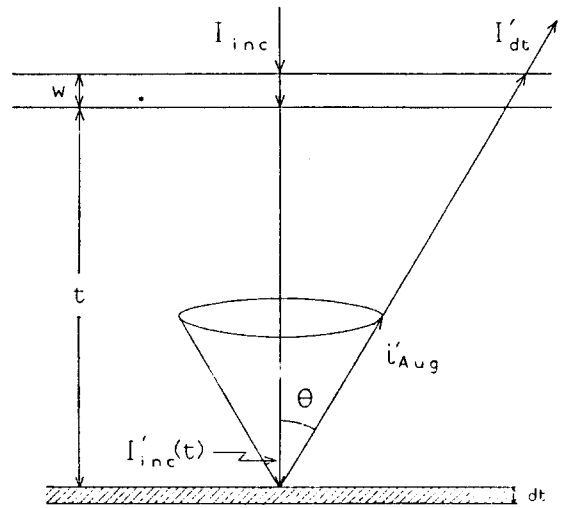

Fig 2. A schematic representation of adsorbed overlayer sample showing quantities used in the text.

the crystal is proportional to the number of excited atoms. If the Auger peak in the N(E) curve is Gaussian, then the peak to peak height of the differentiated Auger spectrum, dN(E)/dE, is also proportional to the emission current.

Since the Auger signals generated below the surface are attenuated exponentially by inealastic scattering as the electrons travel the sample, the relationship between the intensities of the various Auger electron signals as a function of thickness is not linear. From the above statements we can use the relative peak to peak heights to determine the over layer thickness.

2.1. In case of clean single crystal

For the clean sample the intensity, $I_{inc}(t)$, of the incident electrons after penetrating a distance of t as shown in Fig. 1, would be exponentially attenuated by the relation[4]:

$$I_{inc}(t) = I_{inc} e^{-t/\lambda_s(E_i)} \qquad (1)$$

where I_{inc} is the intensity of the incident electron beams, E_i is the incident electron energy, and $\lambda_s(E_i)$ is the IMFP of the incident electron with energy of E_i in the sample. The current, i_{Aug}, due to the Auger electrons produced by the incident electrons in a slab, dt, at depth t can be written as:

$$i_{Aug} = I_{inc}(t) \ P_s \ dt \qquad (2)$$

where P_s is the Auger transition probability of the sample material. Assuming the Auger transition occurred isotropically, the CMA detectable outcoming Auger electron intensity, I_{dt}, on the surface is:

$$I_{dt} = 2\pi \ i_{Aug} \ e^{-t/\lambda_s(E_s)\cos\theta} \qquad (3)$$

where E_s is the Auger electron energy of the sample material, $\lambda_s(E_s)$ is the IMFP of those Auger electrons, θ is the CMA acceptable angle and 2π is the solid angle of the cone. After integrating equation (3) from the surface (t=0) to the bulk (t=∞), the total Auger current intensity, I_s, from the sample will be:

$$I_s = 2\pi \ P_s \ I_{inc}\left[\frac{\lambda_s(E_i) \ \lambda_s(E_s)\cos\theta}{\lambda_s(E_s)\cos\theta + \lambda_s(E_i)}\right] \qquad (4)$$

2.2. In case of deposited overlayers on the single crystal

In case we have overlayers of the thickness w on the substrate as shown in Fig. 2, the intensity, $I'_{inc}(t)$, of the incident electrons after penetrating the overlayers and substrate of depth t will be attenuated by:

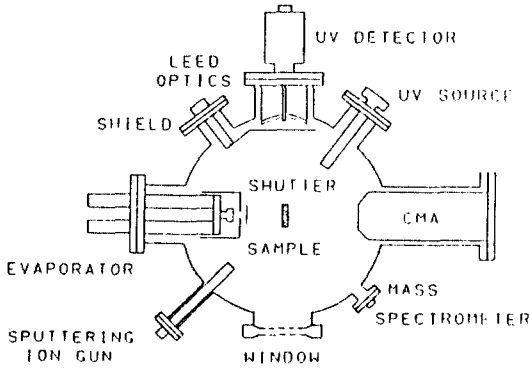$$I'_{inc}(t) = I_{inc} \ e^{w/\lambda_o(E_i)} \ e^{-t/\lambda_s(E_i)} \qquad (5)$$

Fig 3. A schematic diagram of the Varian system.

Fig 4. Auger spectra of copper for various iron cove-rage: (a) clean copper, (b) 0.8 ML iron on cop-per, and (c) 10 ML iron on copper.

where $\lambda_o(E_i)$ is the IMFP of the incident electrons with energy E_i in the overlayer material. After similar integrations, the total detectable Auger current intensity, I'_s, produced from the substrate material can be expressed:

$$I'_s = 2\pi \; P_s \; e^{\;-w\left(\dfrac{\lambda_o(E_s)\cos\theta + \lambda_o(E_i)}{\lambda_o(E_i) \; \lambda_o(E_s)\cos\theta} \right)}$$

$$\left(\dfrac{\lambda_s(E_i) \; \lambda_s(E_s)\cos\theta}{\lambda_s(E_s)\cos\theta + \lambda_s(E_i)} \right) \qquad (6)$$

To obtain the Auger intensity, I_o, due to the overlayer material only, we need to integrate equation (3) from surface (t=0) to overlayer width (t=w) and get the following result:

$$I_o = 2\pi \; P_o \left(\dfrac{\lambda_o(E_i) \; \lambda_o(E_o)\cos\theta}{\lambda_o(E_o)\cos\theta + \lambda_o(E_i)} \right)$$

$$\left(1 - e^{\;-w\left(\dfrac{\lambda_o(E_o)\cos\theta + \lambda_o(E_i)}{\lambda_o(E_i) \; \lambda_o(E_o)\cos\theta} \right)} \right) \qquad (7)$$

Assuming the peak to peak height of the Auger spectrum in the first derivative mode, dN(E)/dE, is proportional to the measurable Auger intensity derived in above equations (6) and (7), we can express the relation between the relative Auger peak height and the overlayer thickness as:

$$\frac{I_o}{I'_s} = \frac{P_o}{P_s} \left\{ \frac{\lambda_o(E_i) \; \lambda_o(E_o) \; [\lambda_s(E_s)\cos\theta + \lambda_o(E_i)]}{[\lambda_o(E_o)\cos\theta + \lambda_o(E_i)] \; \lambda_s(E_i) \; \lambda_s(E_s)} \right\}$$

$$\times \left\{ \frac{1 - e^{\;-w\left[\dfrac{\lambda_o(E_o)\cos\theta + \lambda_o(E_i)}{\lambda_o(E_i) \; \lambda_o(E_o)\cos\theta} \right]}}{e^{\;-w\left[\dfrac{\lambda_o(E_s)\cos\theta + \lambda_o(E_i)}{\lambda_o(E_i) \; \lambda_o(E_s)\cos\theta} \right]}} \right\} \qquad (8)$$
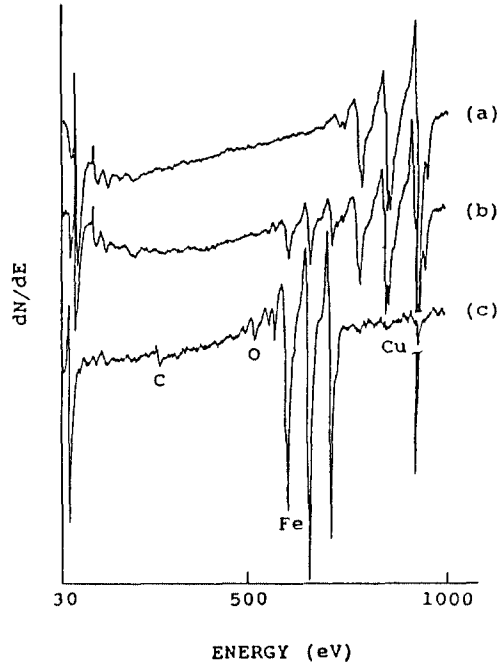
3. Experimental

The experiments were performed in a varian UHV system (Fig. 3) with pressure around 10^{-10} Torr. A near-circular (7 mm diameter $\times$ 1.5 mm thick) high purity copper single crystals were used for the measurments. The crystal direction was verified by the von Laue X-ray back diffraction technique. Mechanical polishing followed by chemical etching produced a clean surface with a bright mirror finish. The sample was then mounted on the sample holder of the manipulator with a chromel-alumel thermocouple spot-welded to the side of the sample. In vacuum, the crystal was cleaned by alternate cycles of argon ion sputtering and heating upto 500℃ until no surface impurities were detected with Auger spectroscopy (Fig. 4). A final annealing at 500℃ for 5 minutes produced a clean surface with a sharp (1×1) LEED pattern.

A normal incident electron gun was used to perform Auger analysis in this work. The sample was positioned at the focal point of the CMA to maximize the elastic peak. This is achieved by transla-
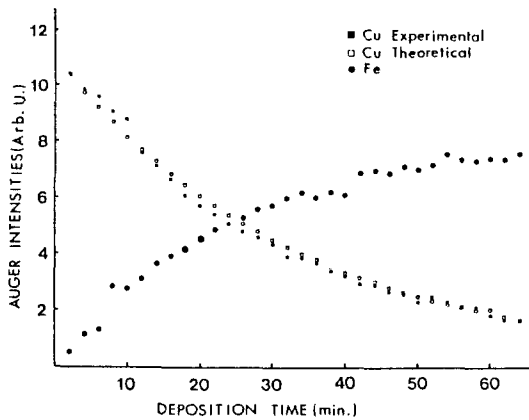
Fig 5. Variation of the Auger intensities (Cu 920 eV and Fe 651 eV) versus deposition time.

Fig 6. Coverage determination using the equation (8) in text. Ratio change (Fe 651 eV/Cu 920 eV) versus monolayers.

ting the sample along the axis of the CMA and by adjusting the focus, x-y deflection, and extractor knobs in Auger gun control module until we get the maximum elastic peak. The beam voltage used was usually 3 keV. Peak to peak voltage of 2 volts was usually used. Scanning width from 30 eV to 1000 eV with 5 minutes sweeping time was used. In the lock-in amplifier 1 mV/sec sensitivity was used. Since the Auger peaks are weak but sharper than the slowly varying background, the Auger spectrum was obtained in the first derivative mode, dN/dE, to enhance the signal. The pure iron source was evaporated from a tantalum Knudsen cell with an alumina boat and deposited in increasing amounts on the copper single crystal surface, while the growth of the 651 eV iron Auger peak and the diminution of the 920 eV copper Auger peak were monitored by depostion time to determine the coverage. During the measurements, the iron deposition source was left on to maintain a constant deposition rate. The sample was rotated to face the evaporator and the CMA alternately. The iron source was interrupted by a shutter when the Auger measurements were made. The formula (8) was used to determine the coverage.

4. Results

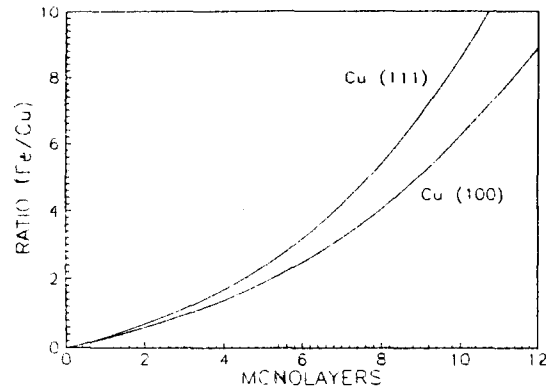The variation in intensity of the 651 eV iron Auger peak and the 920 eV copper Auger peak with

deposition time is plotted in Fig. 5. Initially the intensity of the iron Auger peak increases rapidly with iron coverage; at higher coverage, the rate of increase is diminished. The rate of decrease of the copper Auger peak follows a similar trend.

For a layer-by-layer (Frank-van der Merwe mode)[6] growth, in a continuum approximation, the intensity of the copper substrate peak, I_{CU}, would decrease according to the equation

$$I_{CU} = I_{CU}^{Max} \exp\left(-\frac{l}{d_o}\right) \tag{9}$$

where I_{CU}^{Max} = intensity of a clean copper peak, d_o = mean free path of copper Auger electrons at 920 eV in the iron film, and l = pathlength traversed by the detected electrons. l is related to the thickness of the iron overlayer, d_{Fe}, by $l \cos\theta = d_{Fe}$, where θ is the CMA acceptance angle (42.3°) relative to the surface normal. In Fig. 5, we have also plotted the theoretical curve for the decrease in the intensity of the substrate Auger peak with deposition time. Here we took $d_{Fe} = kt$, where t = deposition time and k = proportionality constant. k was determined by plotting ln (I_{CU}/I_{CU}^{Max}) versus deposition time. The agreement between the theoretical and experimental curves suggests that iron grows approximately layer-by-layer on copper single crystal.

The iron coverage in this study was estimated by the attenuation of the 920 eV copper Auger peak with iron deposition according to equation (8) by

replacing the (P_o/P_s) term by relative sensitivity factors[7] combined with back scattering factors[8-10], θ by 42.3°[11], and substituting the proper values for IMFP terms[1-3]. We can solve the above equation numerically for the overlayer thickness, w, in Angstroms. It is noted that the coverage determination in a monolayer unit depends on the crystal face due to the different distance between layers. The layer distance of the copper (100) direction is 1.805Å while it is 2.086Å for the Cu(111) face. The calculated values of the iron coverage on the Copper (100) and Copper (111) single crystals are plotted in Fig. 6 as a function of the relative Auger peaks ratio (Fe 651 eV/Cu 920 eV) versus monolayers.

5. Conclusion

The Fig. 6 which is based on the equation (8) can be used to determine the overlayer thickness by simply measuring the peak to peak heights of the Auger spectra from the deposited material and substrate one. Even though the above method is not the best one, it is an appropriate way to get the overlayer thickness with ease when we do not have a sophisticated thickness monitor or it does not work properly even we have one. It should be noted that IMFP is the most weight factor in calculating the equation (8) numerically. Also back scattering effect and relative sensitivity factor are important. Those factors are all dependent on electron energy, material and face direction.

References

1. M. P. Seah and W. A. Dench, *Surf Interface Anal.*, **1**, 2 (1979).
2. D. R. Penn, *Phys. Rev. B*, **35**, 482 (1987).
3. S. Tanuma, C. J. Powell, and D. R. Penn, *Surf. Sci.*, **192**, L849 (1987).
4. H. Ibach, Topics in Current Physics Vol. 4, (Springer-Verlag, New York, 1977), Chap. 1.
5. C. J. Powell, *Surf. Sci.*, **44**, 29 (1974).
6. J. H. van der Merwe, *J. Appl. Phys.*, **41**, 4275 (1970).
7. L. E. Davies, N. C. McDonald, P. W. Palmberg, G. E. Riach, and R. E. Weber, Handbook of Auger Electron Spectroscopy 2nd Ed., (Physical Electronics Division, Perkin Elmer Corp., Minnesota, 1976).
8. D. Briggs and M. P. Seah, Practical Surface Analysis by Auger and X-ray Photoelectron Spectroscopy (Wiley, New York, 1983), Chap. 5.
9. S. Ichimura and R. Shimizu, *Surf. Sci.*, **112**, 386 (1981).
10. A. Jablonski, *Surf. Interface Anal.*, **2**, 39 (1981).
11. Varian Vacuum Division, CMA Model No. 981-2607 (1977).