

# Inexpensive Home-Made Single Wavelength Ellipsometer ( $\lambda = 633$ nm) for Measuring the Optical Constant of Nanostructured Materials

L Z Maulana, K Megasari, E Suharyadi, R Anugraha, K Abraha and I Santoso

Departemen Fisika, Universitas Gadjah Mada, Sekip Utara BLS 21 Yogyakarta, Indonesia

Email: iman.santoso@ugm.ac.id

**Abstract.** Inexpensive home-made Single wavelength Ellipsometry with RAE (Rotating Analyser Ellipsometer) configuration has been developed. Spectroscopic ellipsometry (SE) is an optical measurement technique which is based on the measurement of the change of the phase difference ( $\Delta$ ) and the amplitude ratio ( $\psi$ ) between p and s linear polarized of reflected (or transmitted) light. Our RAE configuration system composed of polarizer, sample, analyzer, detector, and He-Ne laser ( $\lambda = 633$  nm) that acted as the monochromatic light source. To test the reliability of our SE system, we measure the optical constant of Au bulk and Cr (30 nm thick) film. The optical constant and the thickness were extracted by employing the pseudo-dielectric function and numerical inversion which is based on the secant method, the  $\psi$  and  $\Delta$  of our SE data which is modelled by Fresnel equation. From the extraction using the secant method we obtain the optical constant of the Au bulk sample with  $n = 0.11$  to  $0.22$  and  $k = 3.26$  to  $3.37$  which is close to that of using pseudo-dielectric method. We obtain the same result for Cr film with  $n = 3.66$  to  $3.81$  and  $k = 5.32$  to  $5.38$  which is close to the result from reference. These results show that our inexpensive home-made Single wavelength Ellipsometry instrument and the extraction method are reliable for determining the optical constant of nanostructured materials.

**Keywords:** Secant methods, optical constant, Ellipsometry

## 1. Introduction

Recently spectroscopy has become a demand at physics, especially on nanostructured material analysis. In the nanoscale, some matters revealed the unique phenomenon compared to the large dimension [1]. Moreover, on some scales, they show the confinement effect [2]. The intrinsic properties of matter can be represented by the complex refractive index. There are several methods to calculate the optical constant of materials that has been developed such as reflectometer and UV-Vis spectroscopy [3]. The determination of both optical constant (refractive index and extinction coefficient) are difficult. This case is due to the correlation and the existence of the imaginary extinction coefficient on the optical constant. Ellipsometry is a technique that both optical constants can be calculated all at once. Ellipsometry is a reliable optical measurement technique that analyzes light reflection (or transmission) from the sample. The main feature of ellipsometry is measured the change in polarization of light upon reflection or transmission from the sample. It is well known as a



very sensitive and nondestructive technique in materials characterization methods. Moreover, it allows to measure accurately and high reproducibility the thicknesses and complex refractive index or dielectric constant of a given materials. In the Ellipsometry measurement, the optical model defining are necessary. That case means any dimension of the sample can be investigated according to our model, even in the nanostructured of the thin film. The precision of Ellipsometry measurement changes proportionally with the wavelength [4,5]. With the light source in this experiment, we can get less than 1 nm precision that leads to the precise result of the optical constant measurement. This paper is explaining the optical constant measurement and calculation of bulky and thin film in nanometer thick.

In the ellipsometry measurement, light are considered as the electromagnetic waves that compose of electrical field  $\mathbf{E}$  and magnetic field  $\mathbf{B}$ . Ellipsometric measurement allows us to quantify the phase difference between  $\mathbf{E}_p$  and  $\mathbf{E}_s$  as  $\Delta$ , and the ratio of their amplitudes given by  $\tan \psi$ . If a sample undergoes a change especially in its thickness or the material composition, its reflection properties will change. For a reflecting surface, the forms of  $\Delta$  and  $\psi$  are defined:

$$\Delta = \delta_p - \delta_s, \quad \tan \psi = \frac{|r_p|}{|r_s|}. \quad (1)$$

where  $\delta_p$  and  $\delta_s$ , are the phase changes for the  $p$  and  $s$  components of light and  $r_p$  and  $r_s$  are the complex Fresnel reflection coefficients. Furthermore, in mathematical language, the ellipsometric analysis is based on Fresnel's equations for polarized light encountering boundaries in the planar sample [5]. The famous ellipsometric equation is conventionally written as:

$$\rho \equiv \frac{r_p}{r_s} \equiv \tan \psi e^{i\Delta}. \quad (2)$$

The general expressions for  $r_p$  and  $r_s$  for the interface between mediums are given by:

$$r_p \equiv \frac{E_{rp}}{E_{ip}} = \frac{N_t \cos \theta_i - N_i \cos \theta_t}{N_t \cos \theta_i + N_i \cos \theta_t} \quad (3)$$

$$r_s \equiv \frac{E_{rs}}{E_{is}} = \frac{N_i \cos \theta_i - N_t \cos \theta_t}{N_i \cos \theta_i + N_t \cos \theta_t}.$$

where the ambient with complex refractive index  $N_i$  and the air with complex refractive index  $N_t$  with  $\theta_o$  and  $\theta_t$  are the angles of incidence and refraction. These expressions can be implied on single and multilayer medium. As long as  $\psi$  and  $\Delta$  are measured at a given wavelength, the optical parameters and thickness sample can be extracted [4].

The ellipsometer has been constructed in RAE (Rotating Analyser Ellipsometry) configuration. Ellipsometry can perform in reflection and transmission mode. Employing  $2 \times 2$  Jones-matrix formalism on RAE configuration, the reflected electric field  $\mathbf{E}_r$  regarding the incident field  $\mathbf{E}_i$  can be expressed in matrix form:

$$\begin{bmatrix} E_A \\ 0 \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} \cos A & \sin A \\ -\sin A & \cos A \end{bmatrix} \begin{bmatrix} \sin \psi \exp(i\Delta) & 0 \\ 0 & \cos \psi \end{bmatrix} \\ \times \begin{bmatrix} \cos P & -\sin P \\ \sin P & \cos P \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix}. \quad (4)$$

The above matrix are consisted by detector, Analyzer, sample, Polarizer and light source respectively. By simplifying this form we obtain Fourier equation with two constant consist in it as follows:

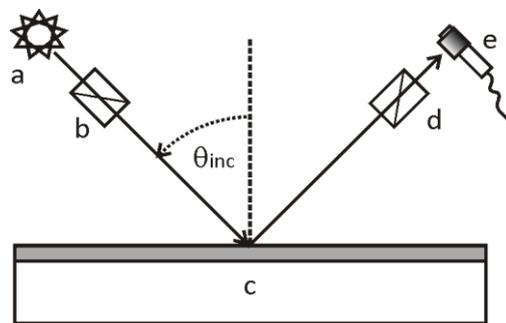
$$I = I_0 [1 + \alpha \cos 2A + \beta \sin 2A] \quad (5)$$

From these two constant  $\alpha$  and  $\beta$ , ellipsometer parameters  $\Delta$  and  $\psi$  can be written as:

$$\tan\psi = \sqrt{\frac{1+\alpha}{1-\alpha}} |\tan\beta|, \quad \cos\Delta = \frac{\beta}{\sqrt{1-\alpha^2}}. \quad (6)$$

## 2. Experimental Method

We describe the instrument and calculation methods of inexpensive home-made single wavelength ellipsometer (cost below \$8000). This ellipsometer composed by the light source, polarizer, sample holder, analyzer, and detector. The light source is a He-Ne laser tube emitting less than 5mW linearly polarized red spectrum ( $\lambda = 633$  nm). Rochon's prism (by Edmund Optic) configuration has been used as the polarizer and analyzer. Those prisms are mounted in vernier scales along  $360^\circ$  vertically with  $1^\circ$  precision. Sample holders were a form of acrylic substance that can be rotated easily along  $360^\circ$  horizontally to determine the incidence angle. The monochromatic light propagated from He-Ne laser polarized by quartz Rochon polarizer through the sample can be seen in Figure 1. The reflected beam passes through another quartz Rochon Analyzer caught by LP Power meter detector. A schematic diagram of these ellipsometers is shown in Figure 1.

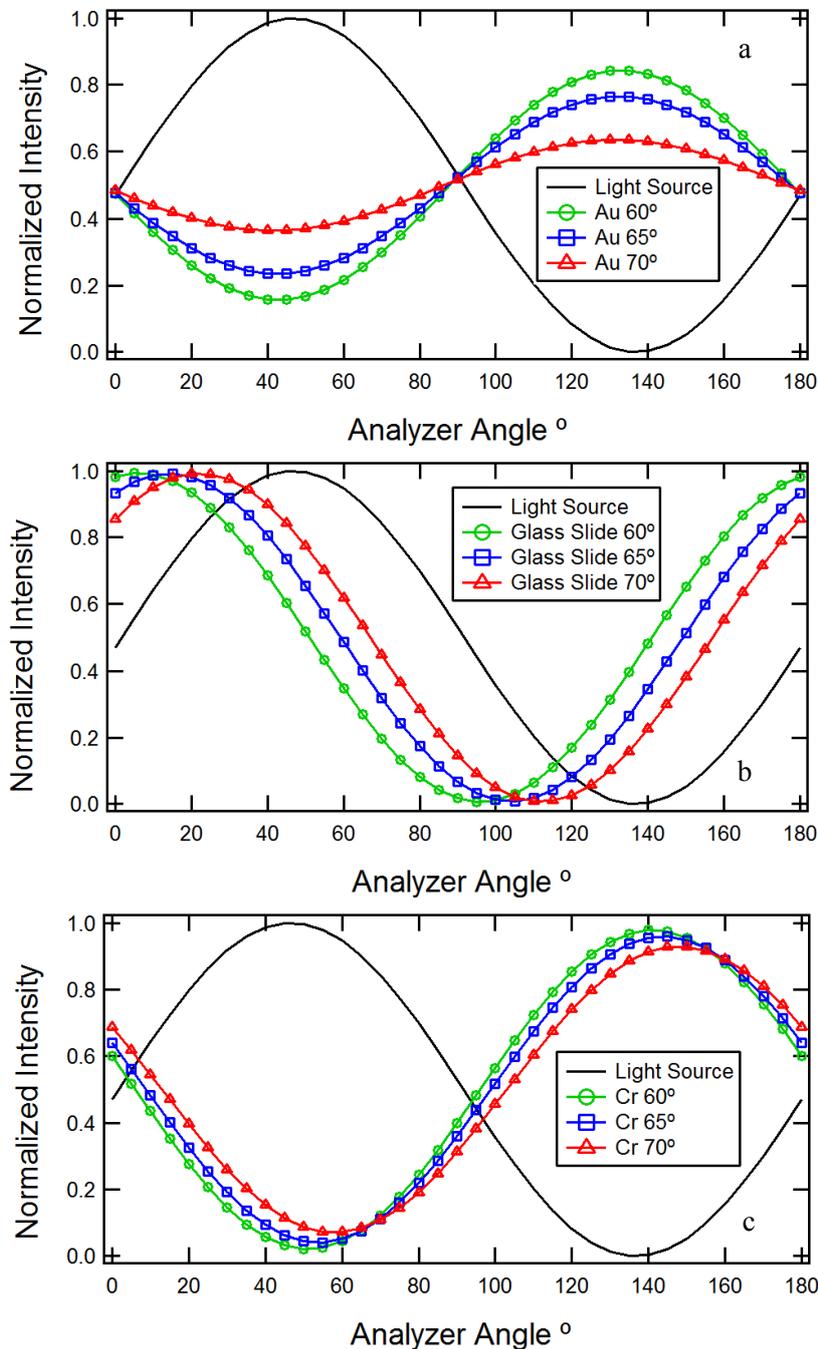


**Figure 1.** Schematic diagram of RAE configuration, (a) monochromatic light source (b) fixed linear polarizer (c) an isotropic sample (d) linear rotated analyzer and (e) photodetector.

In this paper two kinds of materials as the bulk and thin film are investigated. Purchased evaporated Au on a glass substrate is used as the bulk sample. Sputtered Cr with 30 nm thick on a glass substrate is used as the thin film sample. Thin films from deposition usually form on planar with cm dimension depend on the substrate. To fit the sample on the sample holder, the sample must be resized in  $0.3 \times 0.5$  cm  $\times$  1 cm maximum. The sample in out of this range must be fitted with other attachment. The first experimental section begins by setting up the Ellipsometer system. The important thing that should be done in the optical measurement is the instruments alignment. In this case, we made it sure that the light are trough the sample in  $0^\circ$  vertically and horizontally and continue to set the incident angle on  $60, 65^\circ$ , and  $70^\circ$ . An RAE's configuration analyzer are rotated in  $0^\circ$  to  $180^\circ$  with step of  $5^\circ$ . The initial and last intensity were recorded during the measurement and divided to get a normalized intensity versus analyzer angle.

Figure 2 shows intensity versus analyzer angle of Au, glass slide, and Cr 30 nm respectively. For each data analyzer is rotated from  $0^\circ$  to  $180^\circ$  with  $5^\circ$  step. The continuum line guides to light of  $45^\circ$  linear polarized light source, and the other line shows the ellipsometry curve indifference incident angle. As shown in those Figures, reflected light has changed both in amplitude and especially in that phases, that shows the interaction between electromagnetic waves and matter. This paper explains the measurement of the bulk Au (Figure 2a) and the Cr (Figure 2c) thin film. Since both samples are deposited on the glass slide substrate we also show glass slide measurement. Normally the intensity versus analyzer angle in the Ellipsometry measurement are shaped as follow equation 5. All those figures in Figure 2 show the same sinusoidal shape; they have difference peak and close position. Every kind of sample and the specific thickness has a characteristic on the intensity versus analyzer

angle. The special sinusoidal shape that has specific  $\alpha$  and  $\beta$  will lead to one of the intrinsic properties of matter that called the optical constant.



**Figure 2.** Intensity versus analyzer angle of (a) Au, (b) glass slide, and (c) Cr 30 nm.

### 3. Results and discussion

The intensity vs. analyzer angle that is modeled in Eq. 5 were curve fitted with that's equation to obtain  $\alpha$  and  $\beta$ . Those parameters were used to calculate the ellipsometry parameters  $\psi$  and  $\Delta$  with equation 6 and of course  $\rho$  from equation 2. The precision of  $\psi$  and  $\Delta$  can be expressed from the measurement errors of Fourier coefficients following from equation 7 [5].

$$\begin{aligned}\delta\psi &= -\frac{(1 - \cos 2P \cos 2\psi)^2}{2 \sin^2 2P \sin 2\psi} \delta\alpha, \\ \delta\Delta &= -\frac{\cos \Delta (\cos 2P - \cos 2\psi)(1 - \cos 2P \cos 2\psi)}{2 \sin^2 2P \sin \Delta \sin 2\psi} \delta\alpha \\ &\quad - \frac{(1 - \cos 2P \cos 2\psi)}{\sin 2P \sin \Delta \sin 2\psi} \delta\beta.\end{aligned}\quad (7)$$

In this equation  $P$  is polarization angle fixed in  $45^\circ$ . The change of the phase difference ( $\Delta$ ) and the amplitude ratio ( $\psi$ ) can be united in complex form  $\rho$ . The ellipsometry parameters are shown in the Table1 to 3.

**Table 1.** Ellipsometry parameters of Au sample.

Inc. Angle	$A$	$\beta$	$\psi^\circ$	$\Delta^\circ$
$60^\circ$	$-0.050 \pm 0.005$	$-0.683 \pm 0.005$	$43.7 \pm 0.2$	$133.2 \pm 0.4$
$65^\circ$	$-0.0468 \pm 0.0001$	$-0.529 \pm 0.007$	$43.757 \pm 0.001$	$121.9 \pm 0.5$
$70^\circ$	$-0.0340 \pm 0.0005$	$-0.268 \pm 0.002$	$44.13 \pm 0.05$	$105.5 \pm 0.1$

**Table 2.** Ellipsometry parameters of glass slide.

Inc. Angle	$I_0$	$A$	$\beta$	$\psi^\circ$	$\Delta^\circ$
$60^\circ$	0.45	$0.963 \pm 0.004$	$0.206 \pm 0.002$	$82.2 \pm 0.1$	$40 \pm 4$
$65^\circ$	0.46	$0.863 \pm 0.004$	$0.467 \pm 0.008$	$74.8 \pm 0.1$	$22 \pm 4$
$70^\circ$	0.46	$0.707 \pm 0.002$	$0.683 \pm 0.002$	$67 \pm 2$	$14 \pm 1$

**Table 3.** Ellipsometry parameters of Cr thin film.

Inc. Angle	$I_0$	$A$	$\beta$	$\psi^\circ$	$\Delta^\circ$
$60^\circ$	0.46	$0.201 \pm 0.001$	$-0.933 \pm 0.001$	$50.90 \pm 0.03$	$162.34 \pm 0.04$
$65^\circ$	0.46	$0.279 \pm 0.001$	$-0.871 \pm 0.002$	$53.21 \pm 0.09$	$155.2 \pm 0.2$
$70^\circ$	0.46	$0.375 \pm 0.001$	$-0.768 \pm 0.001$	$56.10 \pm 0.03$	$146.04 \pm 0.06$

Basically, the optical constants are calculated by solving the Fresnel equation (Eq. 3). In large shape two media are considered. They are ambient with optical constant  $N_0$  and sample medium with optical constant  $N_1$ . These Fresnel equations are already shown in Eq. 3. The bulk shape sample optical constants are swimmingly calculated with famous pseudo-dielectric function below [4].

$$\hat{\varepsilon} = \hat{n}^2 = \sin^2 \theta_0 \left[ 1 + \tan^2 \theta_0 \left( \frac{1 - \rho}{1 + \rho} \right)^2 \right]. \quad (8)$$

More medium are involved more difficult and complicated equation formed. On the thin film sample, the light can propagate from ambient trough the substrate make three mediums are considered in thin film model. There are film thickness  $d$ , ambient with optical constant  $N_0$ , thin film with optical constant  $N_1$ , and the substrate with optical constant  $N_2$  are involved. The Fresnel equation of thin film shapes are shown below [4]:

$$r_{012,p} = \frac{r_{01,p} + r_{12,p} \exp(-i2\beta)}{1 + r_{01,p} r_{12,p} \exp(-i2\beta)}, \quad r_{012,s} = \frac{r_{01,s} + r_{12,s} \exp(-i2\beta)}{1 + r_{01,s} r_{12,s} \exp(-i2\beta)}, \quad (9)$$

$$\beta = \frac{2\pi d}{\lambda} N_1 \cos \theta_1 = \frac{2\pi d}{\lambda} (N_1^2 - N_0^2 \sin^2 \theta_1)^{1/2}. \quad (10)$$

There are several ways to solve the Fresnel equation. In this paper Secant methods were used to calculate it. Secant methods are the development of Newton methods presented below [6]:

$$x_{k+1} = x_k - \frac{(x_k - x_{k-1})f(x_k)}{f(x_k) - f(x_{k-1})}. \quad (11)$$

As mentioned above  $\psi$  and  $\Delta$  can be transformed to  $\rho_{\text{exp}}$ , and we get  $\rho_{\text{calculation}}$  from Fresnel equation. Secant methods are work on  $\rho_{\text{exp}}$  and  $\rho_{\text{calculation}}$  to be same by putting an error on them. We divine error 0.01 to all of the Secant methods calculation. The function put in Secant methods are depend on the optical model of sample. Equation 12 shows bulky sample and eq. 13 for thin film sample.

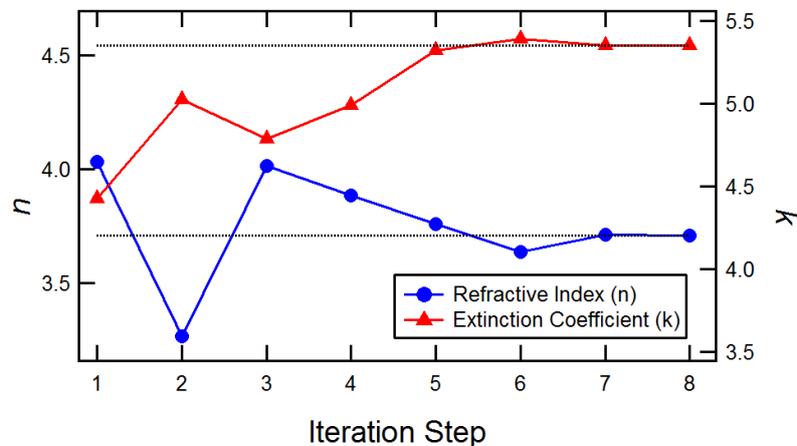
$$f(x) = \rho_{\text{exp}} - \frac{\left( \frac{N_1 \cos \theta_0 - N_0 \cos \theta_1}{N_1 \cos \theta_0 + N_0 \cos \theta_1} \right)}{\left( \frac{N_0 \cos \theta_0 - N_1 \cos \theta_1}{N_0 \cos \theta_0 + N_1 \cos \theta_1} \right)} = 0. \quad (12)$$

$$f(x) = \rho_{\text{exp}} - \frac{\left\{ \frac{r_{01,p} + r_{12,p} \exp(-i2\beta)}{1 + r_{01,p} r_{12,p} \exp(-i2\beta)} \right\}}{\left\{ \frac{r_{01,s} + r_{12,s} \exp(-i2\beta)}{1 + r_{01,s} r_{12,s} \exp(-i2\beta)} \right\}} = 0. \quad (13)$$

As the bulk sample, both ways of secant methods and pseudo-dielectric function calculation of Au are reported. Both calculation methods show the same result that present in Table 4. Its proved that both calculation are reliable to calculated optical constant. We obtain the complex refractive index of the Au bulk sample with  $n = 0.11$  to  $0.22$  and  $k = 3.26$  to  $3.37$  which is close to the result from reference [7,8,9,10]. Basically, the optical constant is the intrinsic properties of matter that shows same value even on the difference angle of incident. There are some discrepancies of our result and the reference. It may come from parallax error, sample tilting and surface inhomogeneity. These results are leading us to developed this algorithm and computational program to calculated another kind of sample.

**Table 4.** Optical constant of Au by secant methods and pseudo-dielectric function calculation.

Inc. Angle	$P$	$\hat{n}$	$\hat{n}$ secant
60°	-0.652804 - 0.696211 i	0.23 + 3.34 i	0.23 + 3.34 i
65°	-0.507264 - 0.812135 i	0.18 + 3.38 i	0.18 + 3.38 i
70°	-0.260148 - 0.934447 i	0.11 + 3.26 i	0.11 + 3.26 i



**Figure 3.** Iteration step of Cr 30 nm optical constant extraction on 65° of incident of angle.

Secant is the popular computational method to calculate non-linear or other complicated equation. In this paper, we perform these methods to extract the complex refractive index of the thin film sample. These methods successfully calculated the optical constant of the bulk sample. All of the secant methods calculation we choose 0.01 as the tolerance value. The reliability of the computational methods to solve some equations can be seen from how fast they reach the tolerance value, and the convergence of that iteration. Figure 3 shows the iteration step sample of our experiment. All of the secant calculation need less than 15 iterations to reach the final step. The convergence iteration in Figure 3 makes sure that these methods are reliable. Table 5 shows the complex refractive index of the Cr thin film with  $n = 3.66$  to  $3.8$  and  $k = 5.33$  to  $5.38$  which is close to the result from reference[10,11]. There are some discrepancies between our result and the reference result, especially on the extinction coefficient. The other reports are explain that deposition methods influence the optical properties of the film due to the crystal structures and the surface roughness [12]. Chromium is belonging to the wet metal that easily oxidized that lead to swallowing this coefficient [11].

**Table 5.** Optical constant of Cr 30nm thick by secant methods calculation.

Inc. Angle	$\rho$	$\hat{n}$ secant
60°	-1.17292 – 0.373358 i	3.81 + 5.33 i
65°	-1.21429 – 0.560096 i	3.71 + 5.35 i
70°	-1.23451 – 0.831445 i	3.66 + 5.39 i

#### 4. Conclusion

The construction and analysis methods of the inexpensive home-made Single wavelength Ellipsometry have been successfully developed. The instrument and the extraction method are reliable for determining the optical constant of bulk and nano-thin film sample. On the Ellipsometry, the light acted as an electromagnetic wave that interacted with matter. We obtained the same result of the Au optical constant that has been calculated by using both Pseudo Dielectric function and Secant method make sure that our Secant methods work properly to solve the two medium Fresnel equations. This capability leads the Secant methods been used to solved Cr nano-thin film that have 3 medium Fresnel Equation. These all systems are successfully to obtain the complex refractive index of Au as a bulky sample and Cr as a nano-thin film sample which is closed to the reference.

## 5. References

- [1] Roco M C Mirkin C A and Hersam M C 2011 Nanotechnology research directions for societal needs in 2020: summary of international study. *J Nanopart Res*, DOI 10.1007/s11051-011-0275-5
- [2] Mitin V, Sementsov D I and Vagidov N Z 2010 *Quantum Mechanics for Nanostructures*, Cambridge University Press UK.
- [3] Potter R F, Reflectometer for Determining Optical Constants *Applied Optics* **4** 53-57
- [4] Tompkins H G and Irene E A *Handbook of Ellipsometry* William Andrew New York
- [5] Fujiwara H 2007 *Spectroscopic Ellipsometry Principles and Applications* John Wiley & Sons Ltd UK
- [6] Klein A and Godunov A 2006 *Introductory Computational Physics* Cambridge University Press UK
- [7] Babar S and Weaver J H 2015 Optical constants of Cu, Ag, and Au revisited *Optical Society of America* **53** 477-481
- [8] McPeak K.M., Jayanti S.V., Kress S.J.P., Meyer S., Iotti S., Rossinelli A., and Norris D.J., 2015, Plasmonic Films Can Easily Be Better: Rules and Recipes. *ACS Photonics* **2**, 326–333
- [9] Olmon R L, Slovick B, Johnson T W, Shelton D, Oh S, Boreman G D and Raschke M B 2012 Optical dielectric function of gold *Physical Review B* **86** 235147
- [10] Rakic' D A, Djuris'ic' A B, Elazar J M and Majewski M L 1998 Optical properties of metallic films for vertical-cavity optoelectronic devices *Journal of Optical Society of America* **37** 5271-5283.
- [11] Johnson P B, and Christy RW 1973 Optical constants of transition metals: Ti, V, Cr, Mn, Fe, Co, Ni, and Pd *Physical Review B* **9** 5056
- [12] Tompkins H G, Baker J, and Convey D 2000 Effect of process parameters on the optical constants of thin metal films *Surface and Interface Analysis* **29** 227–231

## Acknowledgments

Grant of PUPPT, Minister of Research, Technology, and Higher Education, Indonesia, Contract number: 733/UNI-P.III/LT/DIT-LIT/2016.

Nano-Fabrication of Nagoya University, Culture, Sport, Science, and Technology (MEXT) Nano-Project Platform, Japan, 2012-2017