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Citation
Thin Solid Films, 518(22): 6330-6333

Issue date
2010-09-01

Type
Journal Article

URL
http://hdl.handle.net/2298/19818

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Development of thickness measurement program for transparent conducting oxide thin films

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The gallium doped zinc oxide has been one of the candidates for the transparent conducting oxide thin film electrode. It is not suitable to use a conventional light interference method to measure the thickness of the gallium doped zinc oxide thin film because the refractive index and extinction coefficient of the thin film is unknown during the optimization of the deposition conditions. In this paper, we report on the details of the film thickness program which uses the measured optical and electric properties and relationship between the plasma frequency and the optical constant of the film. The obtained film thickness of the prepared gallium doped zinc oxide thin film using the program was comparable with thicknesses measured by a cross-sectional analysis of the atomic force microscopy and the surface profiler. Moreover, the optical constant of refractive index and extinction coefficient of the film could also be estimated.

KEYWORDS: transparent conducting oxide, gallium doped zinc oxide, pulsed laser deposition, film thickness, Drude’s theory, hall effect measurement
1. Introduction

Recently, preparation of transparent conducting oxide (TCO) thin films has attracted much attention for the use as the transparent electrode of flat panel displays, solar cells and so on [1-6]. One of the very important evaluation subjects for the TCO thin film is the film thickness. We have employed a cross-sectional analysis of scanning electron microscopy or atomic force microscopy and a mechanical surface profiler to measure film thickness. On the contrary, non-contact optical measurement technique is very useful since the technique gives no damage on the film. There are several non-contact thickness measurement techniques of a light interference (Peak valley) method, a multiple reflection interference method and a polarization analysis method [7] which includes an ellipsometry. The light interference method is the simplest but needs database of refractive index \( (n) \) and extinction coefficient \( (k) \) then it is not suitable for unknown or non-stoichiometric materials. The multiple reflection interference method needs strong reflection of the film or coating of high reflectance film on the target film. The polarization analysis method can measure not only thickness but also \( n \) and \( k \) of the film. However, this method needs mechanical accuracy then it is relatively expensive.

In this work, we developed a thickness measurement program which uses measurement results of optical and electric properties of the film. The program uses a relationship between the plasma frequency \( (\omega_p) \) and the optical constant of \( n \) and \( k \) of the film. Therefore, it is also possible to estimate wavelength \( (\lambda) \) dependence of the \( n \) and \( k \) of the film by the program. We prepared gallium-doped zinc oxide (GZO) thin films which is one of the candidates of the alternative TCO thin films to indium tin oxide (ITO) on silica glass substrates using pulsed laser deposition and applied the program to measure the film thickness.
2. Flow chart of program

Figure 1 shows a flow chart of the thickness measurement program. We use measured values of transmittance ($T$), reflectance ($R$), hall voltage ($V_H$) and carrier mobility ($\mu_H$) of the film in this program. The $V_H$ and $\mu_H$ are measured by the hall effect measurement. At first, the $\omega_p$ is calculated to lead the $\lambda$ dependence of the $n$ and $k$ of the film. The $\omega_p$ can be expressed by the equation (1) from Drude’s theory using electric properties of carrier concentration ($N$) and relaxation time ($\tau$) of the film.

$$\omega_p = \sqrt{\frac{Ne^2}{\varepsilon \mu \tau m} - \frac{1}{\tau^2}}$$  \hspace{1cm} (1)

Here, the $\varepsilon$ is dielectric constant and the $m$ is effective mass of electron. The $\varepsilon$ of 4 and the $m$ of $2.7 \times 10^{-31}$ kg were used in case of the GZO thin film. However, since the $N$ is a function of $t$ when it is measured by hall effect measurement as shown in equation (2), it is impossible to use the $N$, then the $V_H$ is used as a parameter. Consequently, the $N$ can also be determined after the determination of the $t$. Moreover, the $\mu_H$ measured by hall effect measurement is used to determine the $\tau$ using equation (3). Therefore, $\omega_p$ is calculated from the equation (1) using the equations (2) and (3).

$$N = \frac{IB}{etV_H}$$  \hspace{1cm} (2)

$$\tau = \frac{m\mu_n}{e}$$  \hspace{1cm} (3)

In the equation (2), the $I$ and the $B$ are the current and the applied magnetic field for the hall effect measurement, respectively.

The $n$ and $k$ have relationship between the $\omega_p$ as shown in equations (4) and (5)

$$n^2 - k^2 = \varepsilon \left(1 - \frac{\omega_p^2}{\omega^2 + \tau^{-2}}\right)$$ \hspace{1cm} (4)
We can obtain the $\lambda$ dependence of $n$ by solving these equations as shown in the equation (6).

$$n = \sqrt{\frac{\sqrt{\left(1 - \frac{\omega_p^2}{\omega^2 + \tau^2}\right)^2 + \left(\frac{\omega_p^2}{\omega^2 + \tau^2}\right)^2} + \sqrt{\left(1 - \frac{\omega_p^2}{\omega^2 + \tau^2}\right)^2 + \left(\frac{\omega_p^2}{\omega^2 + \tau^2}\right)^2}}{2}}$$

We can calculate the $\lambda$ dependence of $k$ from the equations (4) and (5). However, the $k$ estimated from the equations (4) and (5) can not express the absorption by the band gap of the film when the region of optical measurement includes ultra-violet region. Then, the measured curves of $T$ and $R$ are used for the determination of the $k$ as shown in the equation (7).

$$k = -\ln\frac{T}{(1-R)} \cdot \frac{\lambda}{4\pi}$$

The $n$ and $k$ are just a function of the $t$. The $\lambda$ dependence of the $n$ and $k$ can be calculated when the $t$ is given. The equations (8) and (9) give the $\lambda$ dependence of the $T$ and $R$ using the $n$ and $k$. It is assumed that the light for the optical measurement penetrates from air (refractive index =1) perpendicular to the film deposited on the glass substrate.

$$T = n^2 \frac{A'^2 + B'^2}{C^2 + D^2}$$

$$R = \frac{A'^2 + B'^2}{C^2 + D^2}$$

Here, $A, A', B, B', C$ and $D$ are given as follows.

$$A = (1-n)(n+n_x)+(k+k_x)k + \left(1+n(n-n_x)-(k-k_x)k\right)\cos \delta - \left[(1+n)(k-k_x)+(n-n_x)k\right] \sin \delta \cdot e^{-2t}$$

$$A' = 4(n \cos \delta - k \sin \delta) e^{-t}$$

$$B = (1-n)(k+k_x)-(n+n_x)k + \left[(1+n)(k-k_x)+(n-n_x)k\right] \cos \delta + \left[(1+n)(n-n_x)-(k-k_x)k\right] \sin \delta \cdot e^{-2t}$$

$$B' = 4(n \sin \delta + k \cos \delta) e^{-t}$$
\[ C = (1+n)(n+n_k) - (k+k_n) + (1-n)(n-n_k) + (k-k_n) \cos 2\delta - [1-n](k-k_n) - (n-n_k)k^2 \sin 2\delta \] \[ D = (1+n)(k+k_n) + (n+n_k) + [1-n](k-k_n) - (n-n_k)k^2 \cos 2\delta + [1-n](n-n_k) + (k-k_n)k^2 \sin 2\delta \] 

Where the \( \delta \) is \( 2\pi nt/\lambda \) and the \( \gamma \) is \( 2\pi kt/\lambda \). The \( n_g \) and \( k_g \) are the refractive index and extinction coefficient of the glass substrate.

After the calculation, the calculated \( T \) or \( R \) is compared with the measured \( T \) or \( R \) using at least square method and the \( t \) is determined when the square error is the smallest.

We developed this program via LabVIEW.

3. Thin film preparation

A KrF excimer laser (Lambda Physik Compex205, maximum energy=650 mJ) was introduced into a stainless chamber through a lens a mirror and a quartz window. Finally, it was focused on GZO target (\( \Phi = 3 \text{ cm}, 5 \text{ wt}\% \text{ Ga}_2\text{O}_3 + 95 \text{ wt}\% \text{ ZnO} \)) with the laser energy density of 2 J/cm² (laser energy =200 mJ) and repetition rate of 10 Hz. Silica glass substrate was placed on a substrate holder. The chamber was evacuated base pressure of \( 10^{-3} \) Pa by a turbo pump and then the deposition was carried out at the pressure and at the room temperature. The distance between the target and the substrate was 28 cm. The representative electric properties of the GZO thin films deposited at the above mentioned conditions are the resistivity of \( 1 \times 10^{-3} \Omega \text{ cm} \), the carrier concentration of \( 1 \times 10^{21} / \text{cm}^3 \) and the mobility of 5 cm²/Vs. We could get the relatively good conductivity and optical characteristics in spite of the low temperature deposition and the in-plane uniformity by employing the longer target-substrate distance [8].

4. Results and discussion

The \( T \) and \( R \) curves of the GZO thin film were measured by a set of Tungsten-Halogen light source (Ocean Optics, DT-MINI), a high-resolution spectrometer (Ocean Optics,
HR4000, 200-1100 nm) and optical fibers which connect the light source and the spectrometer. As for the measurement of the $R$, an Aluminum (Al) mirror (SIGMA KOKI, TFAN-30C05-1) was used as the reference of the reflectance. The program has also a function of the measurement and the data accumulation.

The $V_H$ and $\mu_H$ of the film were measured at $I$ of 0.1 mA and $B$ of 0.28 T by the hall effect measurement system.

Figure 2 shows the calculation result of $T$, $n$, $k$ of the GZO thin film obtained by the program using the measured $T$, $R$, $V_H$ and $\mu_H$. It is revealed that the calculated $T$ curve shows relatively good agreement with the measured curve. The film thickness determined by the program was 170 nm while thicknesses measured by an atomic force microscopy (SII, SPM3800N) and a surface profiler (ULVAC, DEKTAK-3ST) were 140 nm and 190 nm, respectively. Figure 3 shows the relationship of the square error between the calculated $T$ and the measured $T$ versus the given film thickness for the calculation. This result was used to determine the thickness of the GZO thin film (170 nm). It is evident from this figure that the resolution for thickness estimation is as high as several nm which is enough for the thickness detection of transparent oxide thin films. One can confirm two peaks and a valley in the $T$ curve of the Fig.2 because the film is not very thin. The conventional optical interference measurement method can work in this case but some additional fitting technique is needed if the thickness becomes thinner. On the contrary, it is possible to estimate thickness of very thin film in case of our program. Figure 4 shows the calculation result of another sample of very thin GZO thin film. The determined thickness using the program was 60 nm. The thicknesses of the same sample measured by the atomic force microscopy and the surface profiler were 60 nm and 76 nm, respectively. We could confirm the relatively good measurement accuracy of the program even as for the very thin film.
However, we also observe the small difference between the measured $T$ and the calculated $T$. It is considered that one of the reasons is the reflectance of the Al mirror which is used as a reference for the reflection measurement. The reflectance of the Al mirror is approximately 90% and it has somewhat $\lambda$ dependency. We will improve the accuracy of the measurement especially for the reflectance in the next step, which may cause the more accurate thickness measurement.

We suppose that this program is useful to measure the thickness of the film even if the $n$ and $k$ of the film are unknown. In addition, this program can give the $N$ as well as the $\lambda$ dependence of the $n$ and $k$ of the film using the optical measurement and the hall effect measurement systems.

5. Conclusion

The novel thickness measurement program using the measured values of $T$, $R$, $V_H$ and $\mu_H$ was developed via LabVIEW. The measured thickness of the GZO thin film deposited by the PLD method using the program almost agreed with that measured by the atomic force microscopy and the surface profiler even the film is very thin (60 nm). This program is available to determine not only the thickness even if the $n$ and $k$ of the film are unknown but also the $N$ and the $\lambda$ dependence of the $n$ and $k$ of the film.

Acknowledgement

The authors would like to express their thanks to Professor H. Kubota and Professor Y. Nakamura of Kumamoto University for valuable advices and technical support.

References


Fig.1  The flow chart of the thickness estimation program. The measured values of transmittance ($T$), reflectance ($R$), hall voltage ($V_H$) and carrier mobility ($\mu_H$) of the film are used in this program.

Fig.2  The calculation result of $T$, $n$, $k$ of the GZO thin film. (Thicknesses measured by this program, atomic force microscopy, surface profiler are 170 nm, 140 nm, 190 nm, respectively.)

Fig.3  The relationship of the square error between the calculated $T$ and the measured $T$ versus the given thickness which is used to determine the thickness of the GZO thin film (170 nm).

Fig.4  The calculation result of $T$, $n$, $k$ of the GZO thin film. (Thicknesses measured by this program, atomic force microscopy, surface profiler are 60 nm, 60 nm, 76 nm, respectively.)