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Thickness Dependence of Optical Properties of Amorphous Indium Gallium Zinc Oxide Thin Films: Effects of Free-Electrons and Quantum Confinement

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Dielectric function of amorphous indium gallium zinc oxide (a-IGZO) thin films is found to shift significantly with the film thickness, which is attributed to the changes in both the bandgap and electron concentration of the IGZO thin films with the film thickness. The ultrathin films (film thickness < ~20 nm) exhibit a bandgap expansion with reducing film thickness due to the quantum confinement effect; while the thicker films (thickness > ~35 nm) demonstrate the free-electron effect, i.e. the Burstein-Moss shift and increase of free-electrons absorption with increasing electron concentration

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Amorphous indium gallium zinc oxide (a-IGZO) is a transparent semiconductor material that has promising application in next generation flat panel display due to its high electron mobility and high transparency in visible.^{1,2} Although intensive electrical studies have been carried out on the IGZO thin films,³⁻⁶ there is still lack of detailed optical study of the complex dielectric function, one of the fundamental optical properties, of the IGZO thin films. It has been shown in our early work that free-electron concentration of a metal oxide thin film could have a significant impact on the optical properties of the oxide film, in particular, the onset absorption near the band edge.⁷ When the electron concentration reaches $\sim 10^{18}$ – 10^{19} cm⁻³, a semiconductor-to-metal transition occurs, and the conduction band of the oxide semiconductor is partially filled with free electrons, resulting in a bandgap expansion as a result of the Burstein-Moss effect.⁸ The dependence of carrier concentration on film thickness could further complicate the situation.⁹ On the hand, it has also been shown that when a crystalline oxide film scales down to the nanoscale regime (e.g., the film thickness is less than ~20 nm), the nanoscale structures exhibit some unique properties different from their bulk counterparts due to quantum confinement effect.¹⁰ Therefore, it is interesting to study the thickness-dependence of the complex dielectric function of a-IGZO thin films as both the free-electron and quantum confinement effects on the optical properties of the oxide films are dependent of the film thickness.

In this work, we present a study of the evolution of optical properties of a-IGZO thin films with film thickness. The bandgap and complex dielectric function of the IGZO thin films with various film thicknesses have been determined with spectroscopic ellipsometry (SE) based on the Tauc-Lorentz-Drude (TL-Drude) model.⁸ It is observed that the complex dielectric function is strongly dependent of the film thickness. The ultrathin films (thickness < ~20 nm) show a blueshift of both the real and imaginary parts of the complex dielectric function and an expansion of bandgap with decreasing film thickness due to the quantum confinement effect; in contrast, the thicker films (thickness > ~35 nm) exhibit both the Burstein-Moss shift and enhancement of free-electrons absorption with increasing film thickness, which are related to the increase of electron concentration in a thicker film.

Experimental

IGZO thin films with various thicknesses were deposited on a HF-cleaned *p*-type Si wafer. The deposition was carried out by RF magnetron sputtering technique using a IGZO target (In:Ga:Zn mole ratio = 1:1:1). The sputtering was conducted with a RF power of

100 W at room temperature in Ar ambient (background pressure of 3×10^{-3} Torr). The complex dielectric function of the IGZO thin films was determined with spectroscopic ellipsometry (SE) (Woollam VB-250) in the wavelength range of 280–1100 nm. The nominal film thicknesses of the IGZO thin films under study are 5, 11, 20, 35, 50, 60 and 100 nm, respectively. The electron concentration of the thin films was measured with the Hall effect measurement. For the ultrathin films thinner than 35 nm, the Hall effect measurement was not able to yield a reliable value of the electron concentration (which was estimated to be lower than 2×10^{17} cm⁻³); for the thin films thicker than 35 nm, the electron concentration was measured to be in the order of $\sim 10^{18}$ cm⁻³.

Ellipsometric Modeling

Figure 1 shows the SE fittings to the measured ellipsometric angles (Ψ and Δ) of the IGZO film with the thickness of 20 nm for three different incident angles. The SE analysis is based on the four-phase model (i.e., air/surface-roughness layer/IGZO layer/Si substrate). The surface-roughness layer is modeled with the Bruggeman effective medium approximation (EMA)¹¹ as a mixture of the IGZO layer and the air void, as expressed below

$$\frac{\epsilon_{IGZO} - \epsilon_i}{\epsilon_{IGZO} + 2\epsilon_i} f + \frac{\epsilon_{air} - \epsilon_i}{\epsilon_{air} + 2\epsilon_i} (1 - f) = 0 \quad [1]$$

where ϵ_i is the effective complex dielectric function of the surface-roughness layer, f is the volume fraction of IGZO in the surface-roughness layer, ϵ_{air} is the dielectric constant of air void ($\epsilon_{air} = 1$ assumed in all SE analysis), and $\epsilon_{IGZO} (= \epsilon_1 + i\epsilon_2)$, where ϵ_1 and ϵ_2 are the real part and imaginary part of the complex dielectric function, respectively) is the complex dielectric function of IGZO. Here ϵ_{IGZO} is the parameter to be determined from the SE analysis. The complex dielectric function (ϵ_{IGZO}) of the IGZO thin films was determined with SE spectral fitting based on the TL-Drude model.⁸ It has been shown that TL-Drude model can provide an excellent spectral fitting for various metal-doped oxide thin films such as Ga-doped ZnO and Sn-doped In₂O₃.⁸

The TL-Drude model is formulated by combining the Tauc-Lorentz (TL) term¹² and Drude term. The TL term is used to model the band-to-band energy absorption, while the Drude term is used to model the free-electron absorption. The imaginary part (ϵ_2) of dielectric function of the TL-Drude model is described by

$$\epsilon_2(E) = \begin{cases} \frac{A_{TL} E_0 C (E - E_g)^2}{(E^2 - E_0^2)^2 + C^2 E^2} \cdot \frac{1}{E} + \frac{A_D \Gamma_D}{E^3 + \Gamma_D^2 E}, & \text{for } E > E_g \\ \frac{A_D \Gamma_D}{E^3 + \Gamma_D^2 E}, & \text{for } E < E_g \end{cases} \quad [2]$$

where (A_{TL} , C) and (A_D , Γ_D) are (oscillator strength, broadening parameters) for the TL and Drude terms, respectively. E_0 is the peak transition energy, and E_g is the bandgap energy. The real part (ϵ_1) of

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Figure 1. SE spectral fitting of the 20 nm IGZO thin film based on the TL-Drude model for the angles of incidence of 65°, 70° and 75°.

the dielectric function of the TL-Drude model can be obtained by the Kramers-Kronig integration of ϵ_2 .

The optical constants including the refractive index (n) and extinction coefficient (k) of the IGZO thin films can be calculated from the value of dielectric function (ϵ_1, ϵ_2) with the following formulas

$$n = \sqrt{\frac{\sqrt{\epsilon_1^2 + \epsilon_2^2} + \epsilon_1}{2}} \quad [3]$$

$$k = \sqrt{\frac{\sqrt{\epsilon_1^2 + \epsilon_2^2} - \epsilon_1}{2}}$$

where (ϵ_1, ϵ_2) are the real and imaginary parts of dielectric function, respectively.

Discussion

Figure 2 shows the complex dielectric function of the IGZO thin films with various thicknesses. The calculated optical constants (n, k) of the IGZO thin films are shown in Fig. 3. As shown in Fig. 2, the film thickness has a significant impact on the dielectric function of the IGZO thin films. It has been reported that when the thickness of ZnO and Ge thin films scales down to 20 nm, the ultrathin films exhibit an expansion of bandgap and a reduction of dielectric function due to the quantum confinement effect.^{10,13} As shown in Fig. 2a for the IGZO thin films thinner than ~ 20 nm, the ultrathin films exhibit a blueshift in both real and imaginary parts of dielectric function with decreasing film thickness. The blueshift indicates an expansion of bandgap, which can be attributed to the quantum confinement effect.¹⁴ As discussed earlier, the ultrathin IGZO films (thickness $< \sim 20$ nm) has the electron concentration of lower than $\sim 2 \times 10^{17} \text{ cm}^{-3}$, which is far below the Mott critical density (10^{18} – 10^{19} cm^{-3}) to observe the BM effect.¹⁵ Therefore, the observed blueshift of dielectric function shown in Fig. 2a is mainly attributed to the quantum confinement effect, which leads to a bandgap opening. Unlike the nanocrystalline ZnO thin film where the electrons are confined in a three dimensional environment

Figure 2. Real part (ϵ_1) and imaginary part (ϵ_2) of the complex dielectric function of the IGZO thin films with various thicknesses. The arrow indicates the increase of film thickness.

Figure 3. (a) Refraction index (n) and extinction coefficient (k) of the IGZO thin films with various thicknesses.

(e.g., nanocrystalline grain size),¹⁰ ultrathin amorphous IGZO films could exhibit the confinement of electrons along the thickness direction (i.e., the one-dimensional quantum confinement (ODQC) effect). It has been shown that thickness dependence of band gaps of ultrathin amorphous materials can be described by the ODQC effect.^{13,16}

Figure 4 shows the bandgap energy of the IGZO thin films yielded from the SE modelling. A significant dependence of bandgap on film thickness is observed. As shown in the figure, the ultrathin films ($< \sim 20$ nm) exhibit an expansion of bandgap energy with decreasing film thickness, which can be mainly attributed to the ODQC effect in the ultrathin amorphous IGZO films. On the other hand, as the electron concentration increases with the thickness and reaches the order of $\sim 10^{18} \text{ cm}^{-3}$, the thicker film (> 35 nm) exhibits a significant expansion in bandgap energy with increasing film thickness. It has been shown that when electron concentration in an oxide film exceeds the Mott critical density (e.g., 10^{18} – 10^{19} cm^{-3} in ZnO), a semiconductor-to-metal transition occurs, and filling of the conduction band with free electrons leads to a bandgap expansion as a result of the BM effect.^{15,17} As shown in Fig. 4, a significant increase of the bandgap energy is observed for the IGZO thin films thicker than ~ 35 nm. The increase of the bandgap energy with increasing film thickness can be explained by the BM effect as a result of a higher electron concentration in a thicker IGZO film. In other words, for the thicker films, the BM effect is dominant while the ODQC effect is insignificant. The explanation of the bandgap expansion by the BM effect is consistent with the large increase of the plasma energy with film thickness as discussed below.

As indicated by Eq. 2, the value of the parameter A_D of the Drude term for free electrons can be obtained from the SE modeling. In the Drude theory, A_D is expressed by⁸

$$A_D = \epsilon_\infty E_p^2 \quad [4]$$

where ϵ_∞ is the high frequency dielectric constant and E_p is the plasma energy. E_p is defined by

$$E_p = \left(\frac{\hbar^2 e^2 N_f}{m^* \epsilon_\infty \epsilon_0} \right)^{1/2} \quad [5]$$

where $\hbar$, e , m^* , ϵ_0 , and N_f are the reduced Planck constant, electron charge, electron effective mass, free space permittivity and free electron concentration, respectively. The plasma energy can be calculated with Eq. 4 by using $\epsilon_\infty = 1$ and the A_D values yielded from the SE fittings. Figure 5 shows the plasma energy of the IGZO thin films as a function of film thickness. As can be observed in the figure, the plasma energy increases with increasing film thickness. The increase of E_p indicates an increase of free-electron concentration in the IGZO films as E_p is proportional to $\sqrt{N_f}$. The large dependence of carrier concentration on film thickness could be attributed to the improvement of polycrystalline grain structures with increasing film thickness.^{18–20} A large increase of electron concentration could significantly influence

Figure 4. Band gap energy of IGZO thin films as a function of the film thickness.

Figure 5. Plasma energy (E_p) of the IGZO thin films as a function of the film thickness.

the band-to-band absorption in the film. As the concentration reaches the order of $\sim 10^{20} \text{ cm}^{-3}$, the modification of electronic states results in a bandgap renormalization due to the mutual exchange and coulomb interactions between the free electrons, and leads to narrowing of the bandgap.²¹ As shown in Fig. 5, the plasma energy (E_p) of the IGZO thin film with the thickness of 100 nm is $\sim 1.11 \text{ eV}$, which is much larger than the value (0.82 eV) of the 50 nm film, showing a large increase of the electron concentration. However, the 100 nm film does not exhibit a large increase in the bandgap energy as compared to the 50 nm film, as shown in Fig. 4. This indicates that in the thicker thin films with a high electron concentration, the bandgap change should

Figure 6. Imaginary part (ϵ_2) of the complex dielectric function of the IGZO thin films calculated with Eq. 2 in the energy range of 0.5–1.5 eV where the free-electron absorption is significant. The arrow indicates the increase of film thickness.

be considered as the net result of the competition between the BM shift and the bandgap renormalization.

Figure 6 shows the imaginary part (ϵ_2) of the complex dielectric function of the IGZO thin films calculated with Eq. 2 in the energy range of 0.5–1.5 eV where the free-electron absorption is significant. It is observed in the figure that ϵ_2 increases with film thickness. This indicates an increase of free-electron absorption in a thicker film due to the increase of electron concentration, which agrees with the increase of E_p with film thickness.

Conclusions

The complex dielectric function and bandgap energy of amorphous indium gallium zinc oxide thin films with various thicknesses have been determined with spectroscopic ellipsometry based on the Tauc-Lorentz-Drude model. Evolution of both the dielectric function and bandgap with film thickness has been observed, which is explained in terms of the quantum confinement effect and the free-electron effect (i.e., the Burstein-Moss shift and free-electron absorption). For the ultrathin films (i.e., the thickness is smaller than $\sim 20 \text{ nm}$), the confinement effect is dominant; however, for the thin films thicker than $\sim 35 \text{ nm}$, the free-electron effect is dominant due to the high electron concentration in the oxide films.

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