Depolarization correction method for ellipsometric measurements of large grain zinc-oxide films

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Introduction

Spectroscopic Ellipsometry (SE) is commonly used to measure the thickness and optical properties of thin layers and has been successfully applied to zinc-oxide (ZnO) samples [1, 2]. However, the evaluation of ellipsometric measurement can be complex on depolarizing samples. Inhomogeneous layer-thickness and scattering are two possible sources of depolarization. Our aim was to investigate these two sources of depolarization and to give a method for handling scattering-caused depolarization.

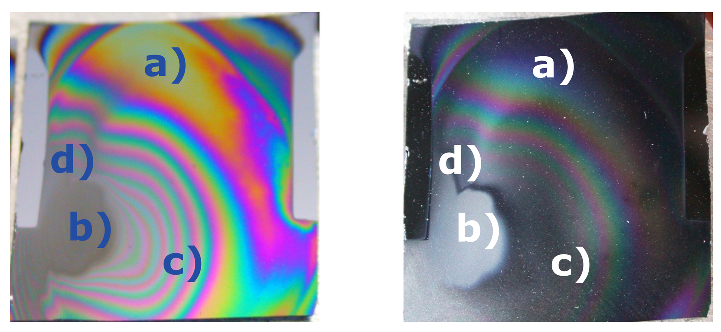
Experimental

ZnO films were produced by Pulsed Laser Deposition (PLD). A pressed ZnO target was ablated by an ArF excimer laser in 1 Pa oxygen background. The ZnO layer was deposited from the laser plume on a resistively heated silicon substrate (600°C was measured in the middle by radiation pyrometer). The regions closest to the current junctions experienced higher current density and therefore higher temperature.
Morphology of the films was characterized with scanning electron microscopy (SEM).
A rotating compensator spectroscopic ellipsometer (Woollam M-2000F) was used to measure the Ψ and Δ values, depolarization of the samples, and Mueller-matrix elements in the 275-1000 nm (1.24-4.5 eV) range. Measurements were performed by using a focused light beam at 65° and 75° angles of incidence.
Investigations were made on

a) **smooth** and **uniform** films,
b) samples with **scattering surface**, where the **film thickness is uniform**,
c) **smooth** samples with **non-uniform** film thickness, and
d) samples on which **both scattering** and **thickness inhomogeneity** appear.

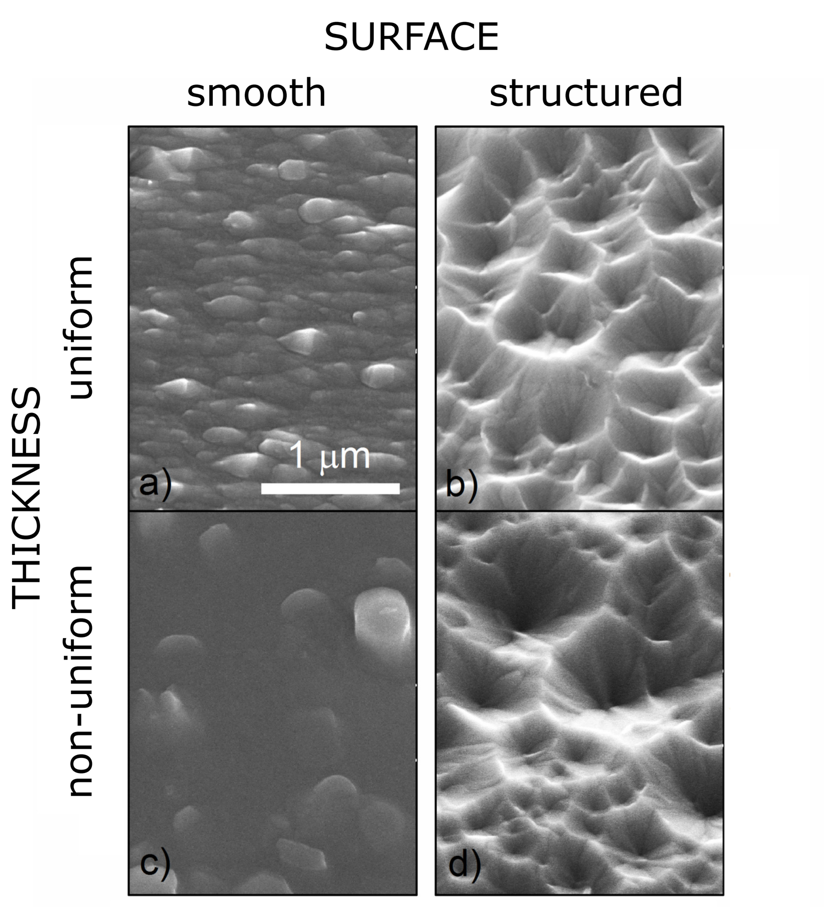
Results

Morphology



Bright-field and dark-field photographs were taken with a digital camera by illuminating the samples with fluorescent lamp and sunlight.

SEM images: the different regions of the ZnO films showed different morphologies.



The PLD technique combined with the resistive heating caused **various morphology and thickness-distribution** of the ZnO film.

Ellipsometry

Mueller-matrices:

Depolarization of a sample can be calculated from Mueller-matrix elements using the following equation [3]:

$$D = 1 - \left(M_{12}^{\ 2} + M_{33}^{\ 2} + M_{34}^{\ 2}\right)$$

Mueller-matrix of a sample acting as a **simple depolarizer** can be defined in the following way [3, 4]:

$$\underline{\underline{M}}^{D} = \beta \cdot \underline{\underline{M}} + (1-\beta)\cdot \underline{\underline{D}} = ((R-1)\cdot\beta + 1)\cdot \begin{bmatrix} 1 & -B\cos 2\Psi & 0 & 0 \\ -B\cos 2\Psi & B & 0 & 0 \\ 0 & 0 & B\sin 2\Psi\cos\Delta & B\sin 2\Psi \sin\Delta \\ 0 & 0 & -B\sin 2\Psi\sin\Delta & B\sin 2\Psi\cos\Delta \end{bmatrix}$$

where β is the fraction of polarized light in the light beam, R is the power reflectivity, $\underline{\underline{M}}$ denotes the Mueller-matrix of an isotropic reflecting sample, $\underline{\underline{D}}$ is the Mueller-matrix of an ideal depolarizer and B is the following constant:

$$B = \frac{R\cdot\beta}{(R-1)\cdot\beta + 1}$$

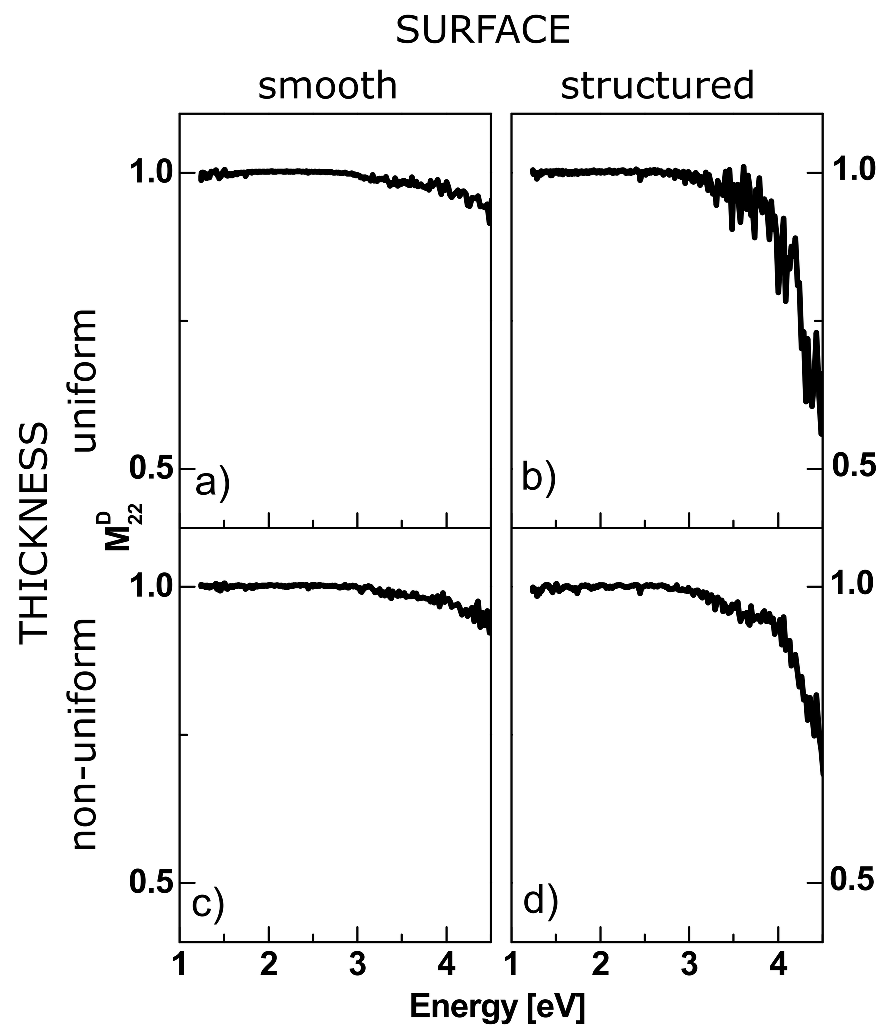
Depolarization caused by **inhomogeneous layer-thickness** can be taken into account by averaging the Mueller-matrices at given thicknesses in the whole thickness range [3]:

$$\underline{\underline{M}} = \langle R\rangle \begin{bmatrix} 1 & -\cos 2\langle\Psi\rangle & 0 & 0 \\ -\cos 2\langle\Psi\rangle & 1 & 0 & 0 \\ 0 & 0 & \sin 2\langle\Psi\rangle\cos\langle\Delta\rangle & \sin 2\langle\Psi\rangle\sin\langle\Delta\rangle \\ 0 & 0 & -\sin 2\langle\Psi\rangle\sin\langle\Delta\rangle & \sin 2\langle\Psi\rangle\cos\langle\Delta\rangle \end{bmatrix}$$

Comparing the two matrices:

M_{22} matrix-element is not altered by thickness non-uniformity, but mainly depends on the fraction of polarized light in the light beam.

Measured M^{D}_{22} values:



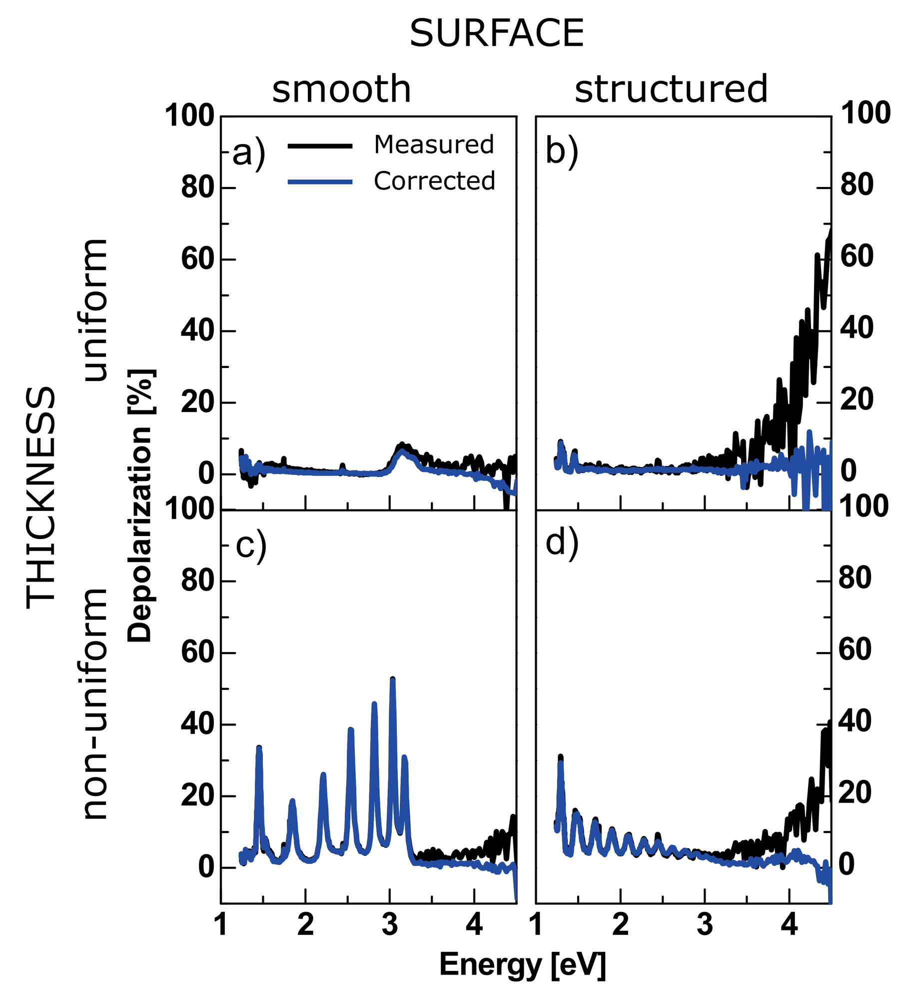
The band gap of ZnO thin film is 3.3 eV [5]. Above this photon-energy the layer is not transparent, there are no more reflections from film-substrate interface. **Non-uniform layer thickness** does not cause depolarization in this region. Therefore in the absorbing region the appearing depolarization is caused by scattering due to comparable UV wavelength and structure size. This type of depolarization can be eliminated by using Eq. (1).

Making the following correction, **the depolarization due to scattering can be eliminated** from depolarization spectrum:

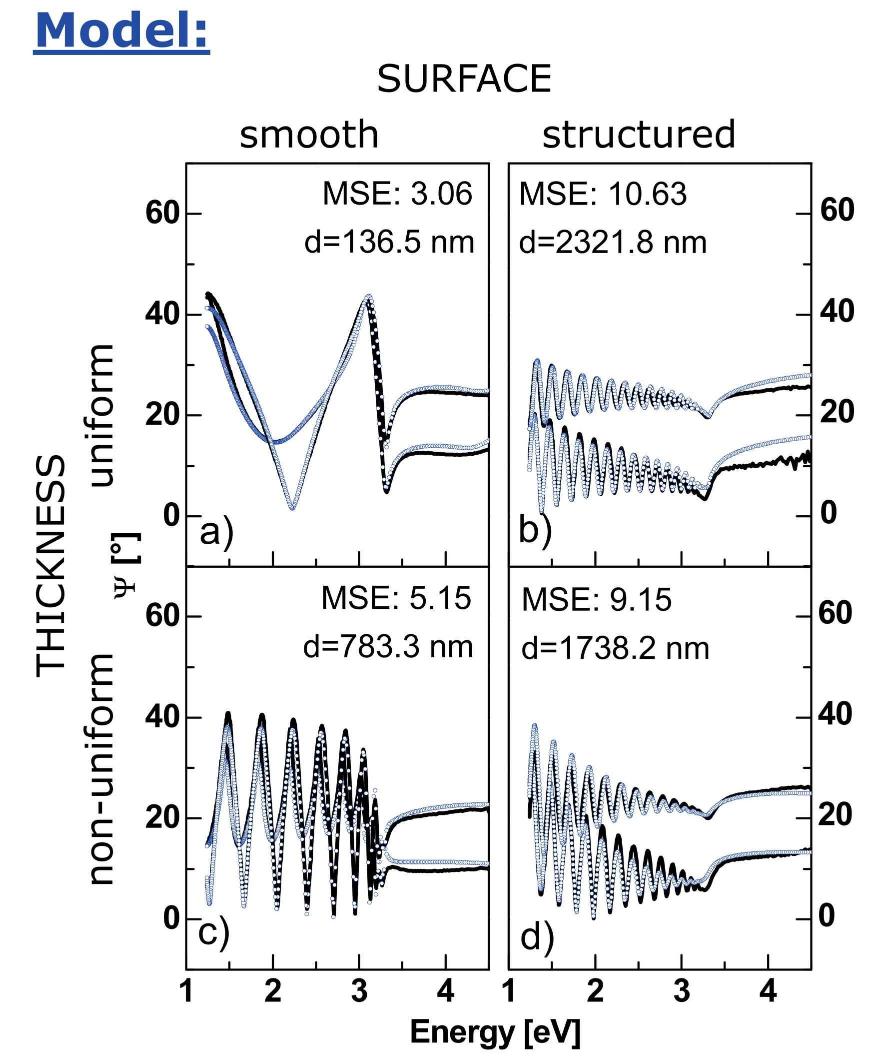
$$D_{corr} = 1 - \frac{M^{D}_{12}{}^{2} + M^{D}_{33}{}^{2} + M^{D}_{34}{}^{2}}{M^{D}_{22}{}^{2}} \qquad (1)$$

If one corrects the Mueller-matrix elements with this type of depolarization, the Ψ and Δ values remain the same because of their definition.

$$\Psi = \frac{1}{2}\cdot\tan^{-1}\left(\frac{\sqrt{M_{33}^{\ 2} + M_{34}^{\ 2}}}{M_{12}}\right); \quad \Delta = \tan^{-1}\left(\frac{M_{34}}{M_{33}}\right)$$



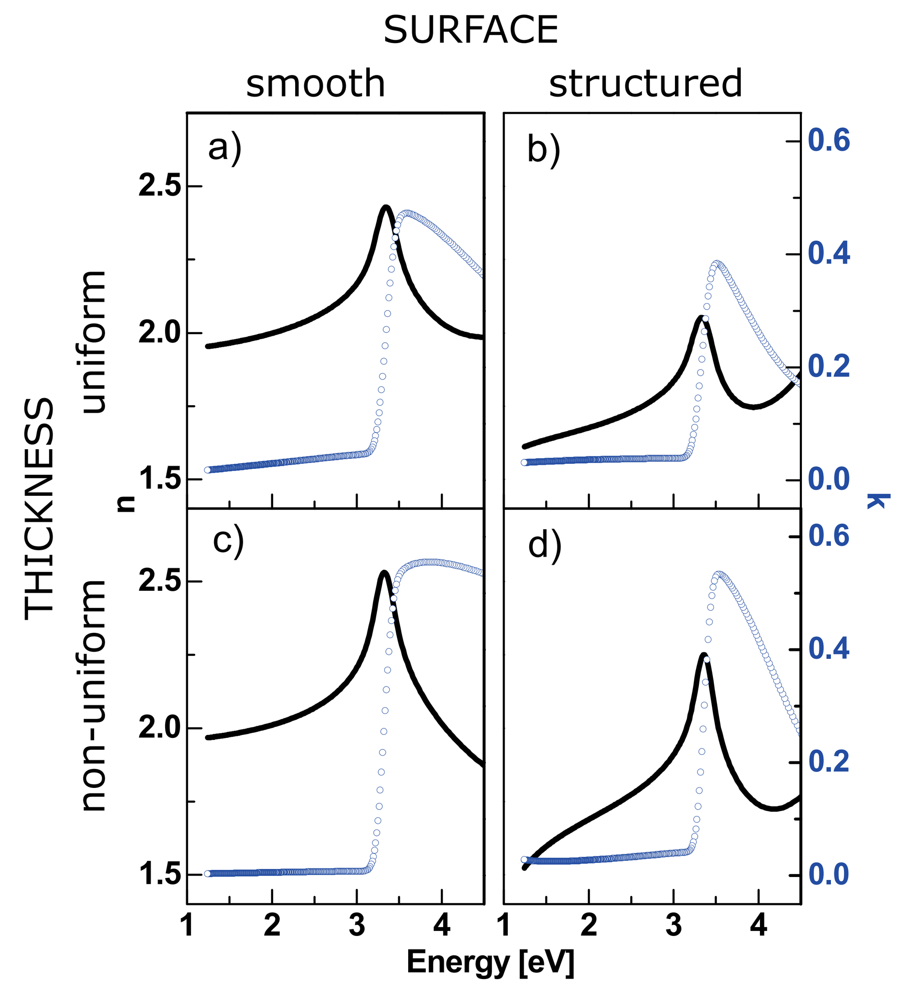
Model:



The absorption edge of ZnO which determines the main characteristics of the measured Ψ and Δ spectra could be described by applying psemi M0 [6] model. Slight absorption in the visible and in the infrared region was modeled with Lorentz and Drude oscillators.

Optical properties:

The *n* and *k* curves of the ZnO layer calculated from the applied model are in accordance with typical ZnO data [1]. **Layer structures** cause **lower refractive index.** Refractive index decrease is related to larger grain size [7], which can be obtained at scattering areas due to higher substrate temperature close to the current junctions.



Conclusions

Mueller-matrix analysis shows that the scattering-caused depolarization can be decoupled from the depolarization appearing because of inhomogeneous layer thickness.
The theoretical description and our measurements demonstrated that depolarization induced by scattering is correlated with M^{D}_{22} matrix-element. If a corrected depolarization is introduced by Eq. (1), then the depolarization curve will show only the effects of inhomogeneous layer thickness, which can be modeled.
Ψ and Δ values are not altered by this correction, but our ellipsometric model can be improved if the corrected depolarization is used in the fitting procedure.

References

[1] H. Yoshikawa, S. Adachi, Jpn. J. Appl. Phys. 36 (1997) 6237.
[2] L. Miao, S. Tanemura, M. Tanemura, S. P. Lau , B. K. Tay, J Mater Sci: Mater Electron (2007) 18:S343–S346.
[3] H. G. Tompkins, E. A. Irene (Eds.), Handbook of Ellipsometry, William Andrew Inc, Springer (2005) p. 65-66., 289-293.
[4] J.-Th. Zettler, Th. Trepk, L. Spanos, Y.-Z. Hu and W. Richter, Thin Solid Films, 234 (1993) 402.
[5] Ü. Özgür et al, JOURNAL OF APPLIED PHYSICS, 98 (2005) 041301.
[6] Guide to Using WVASE32, J. A. Woollam Co., Inc. (2010)
[7] R.G. Heideman, P.V. Lambeck, J.G.E. Gardeniers, Optical Materials 4 (1995) 741-755.

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