Bulk and surface inhomogeneity of r.f. sputtered ITO films: an ellipsometric study

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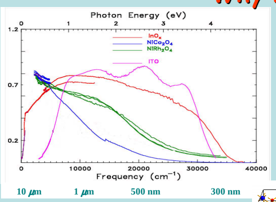
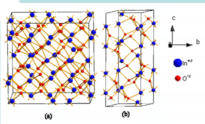
Abstract. *Tin-doped indium oxide (ITO) films have been prepared by r.f. sputtering technique in pure Ar and Ar+O2 atmospheres. Additional substrate heating during the deposition was used for a half of the samples. The influence of both deposition conditions and post-annealing treatment on film inhomogeneity was studied using multiangle spectroscopic ellipsometry. The structural and microstructural properties of the ITO films have been compared to the film inhomogeneity values.*

Why ITO?



The outstanding combination of optical and electrical properties achievable only for this material distinguishes the ITO among other TCOs.

Need of deeper understanding ITO growth mechanisms and the relationship between the material characteristics and the deposition conditions, as well as type of post-deposition treatments

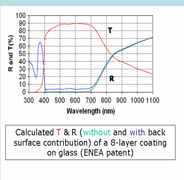


Examples of ITO applications

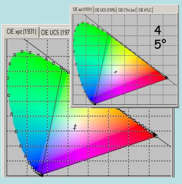
Our Start Point: Art protection coatings

Glass reflection

Color fading

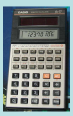


- Blocking λ<410nm
- Attenuation λ >700nm
- AR 410-680nm
- no color alterations



Portable Electronics with Displays

Solar Cells and other PV devices



For many applications relatively thick ITO films are needed.

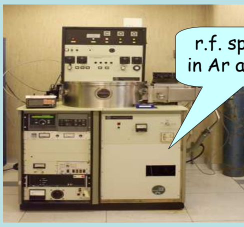
There was not yet published any ellipsometric study for single ITO film thicker than 300 nm ca, nor systematic investigation on ITO film internal formation and re-organization upon post-deposition treatments.

References

A. Krasilnikova Sytchkova, M.L. Grilli, S. Boycheva and A. Piegari, “Optical, electrical, structural and microstructural characteristics of r.f. sputtered ITO films developed for art protection coatings”, Appl. Physics A – Matter, A 89 (2007), 63-72.

R.A. Synowicki, “Spectroscopic ellipsometry characterization of indium tin oxide film microstructure and optical constants”, Thin Solid Films 313-314 (1998), 394-397

Sample preparation and characterization



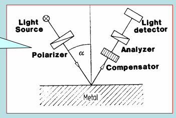
r.f. sputtering in Ar and Ar+O_2

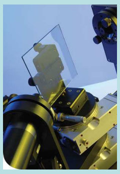
The deposition conditions are optimized to obtain high transparency in the visible and significant reflectance in the NIR.

Sample	P_{O2} (%)	T_{sub}. (°C)	Thickness (nm)	ρ (10^{-4}, Ω cm)	Relative intensity ($I_{(400)}/I_{(222)}$)	Strongest peak at grazing angle XRD
AM1	/	20	1062	4.89	1.77	(222)
AM1a			1216	9.24	1.63	(222)
AM2	/	190	1069	4.06	3.32	(400)
AM2a			1200	5.76	1.54	(222)
AM3	0.6	20	1132	4.64	3.93	(400)
AM3a			1264	6.70	1.91	(400)
AM4	0.6	190	1151	4.60	11.23	(400)
AM4a			1151	5.18	3.52	(400)
AM5	2.8	20	1188	14.85	3.92	(400)
AM5a			1202	51.69	3.93	(222)
AM6	2.8	190	1127	9.35	13.35	(222)
AM6a			1144	11.55	2.43	(400)

Deposition conditions and main characteristics of AM films: a = annealed; P_{O2} – partial oxygen pressure, Tsub – substrate temperature at deposition start, ρ - film resistivity

Variable Angle Spectroscopic Ellipsometry





J.A. Woollam Company VASE ellipsometer

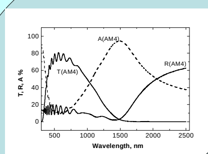
RAE + AutoRetarder Technology
Rotating Analyzer Ellipsometers (RAE)

In reflection mode.
Incidence angles from 50° to 75° with the step of 5°.
Spectral range 320-1700 nm with the step of 10 nm.

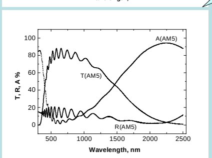
AutoRetarder accurately measures:

- *Ψ and Δ over the full range!*
- *Generalized (anisotropic) Ellipsometry*
- *Depolarization data*
- *Mueller-matrix data*

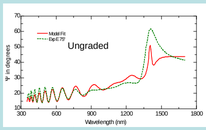
Results

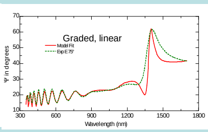


Two ITO films with different position of plasma resonance frequency of free carriers.

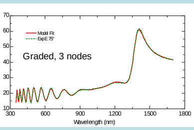


Characterization example: AM4 sample





Model	Mean Squared Error	ITO film thickness (nm)	Surface roughness thickness (nm)
Ungraded	62.2	925	9.0
Graded, linear	47.7	950	9.5
Graded, 3 nodes	9.5	1030	12.7



Analysis of ellipsometric data

- Initially, the multi-angle spectral data were fitted in the frame of a “dispersive homogeneous” model, where a film has plane-parallel boundaries and its material has homogeneous distribution of the refractive index and extinction coefficient along the film thickness. At this step the layer material dispersion was simulated by using an oscillator-based approach which automatically ensures the Kramers-Krönig consistency.
- Two-parametric Drude oscillator was used to reproduce the free carrier absorption in the NIR, while a three-parametric Lorentz oscillator was employed to simulate both absorption and dispersion in the UV. Fitting parameters were amplitude (A_d) and broadening (B_d) for Drude oscillator, while central energy (E_l), amplitude (A_l) and broadening (B_l) were the three fitting parameters of Tauc-Lorentz oscillator.
- The optical model was further refined by adding a surface roughness layer on top of the ITO film.
- In most of the cases, the simple homogeneous model was not able to reproduce the experimental data, in particular at higher wavelengths. Models were then refined introducing inhomogeneity (grading) of the material optical properties along the film thickness.
- The linear grading helped to improve the agreement between experimental and simulated data but the overall fit quality was still quite poor. Therefore, a further flexibility was introduced in the model by using different nodes (up to four) along the film thickness, thus letting the optical constants to vary in a non-linear way.

Conclusions

- VASE is a powerful tool for non-destructive analysis of such complex optical structures like 1 micrometer thick ITO films are. When acquired at vast spectral and angular range, the measured change of polarization state of the incident light, induced upon interaction with the sample, yields a detailed description of the depth profile of a transparent coating.
- A complex and careful modelling is needed, however, in order to obtain an adequate fit simultaneously for all measurement data input, remaining in the frame of a model chosen for a given sample, i.e. using the same set of the fitting parameters. A strict sequence of steps in modelling is required in order to obtain physically meaningful results.
- An interesting correlation between film inhomogeneity studied by VASE and film structure obtained from XRD measurements was observed. The refractive index and extinction coefficient profiles, together with prevalent orientation of film polycrystals, are consequent to substrate heating during the film deposition, and undergo concurrent changes with film re-organisation upon post-deposition annealing.