BEAM REQUEST at Bucharest TANDEM 3MV

Experiment Title **RBS analysis of doped ZnO thin films grown by pulsed electron beam deposition**

Experiment Responsable:

INFLPR: Nume* NISTOR Magdalena (INFLPR) **E-mail address* mnistor@infim.ro**
Phone+4021 4574490

IFIN-HH: Dr. M. GUGIU, Dr. D. DUMITRIU, Dr. D. PANTELICA

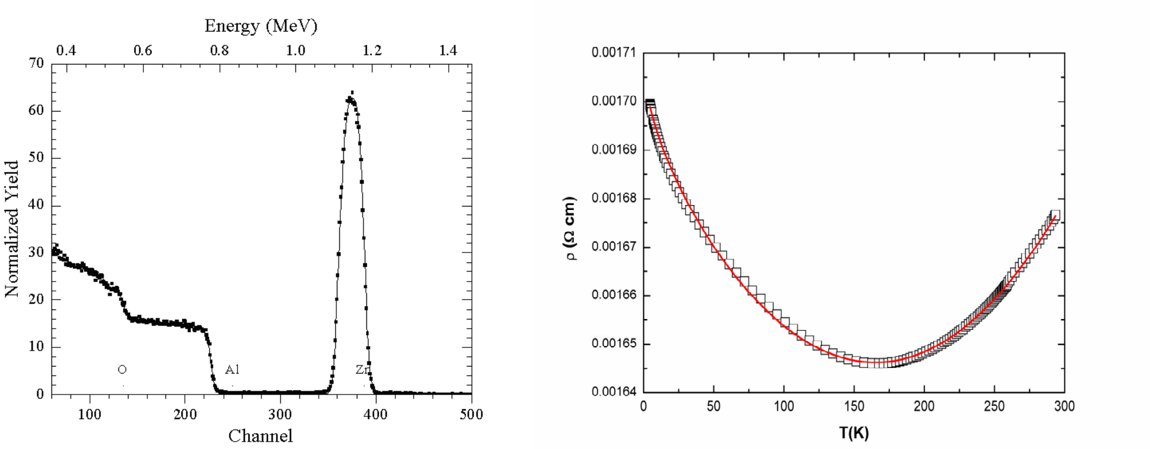
Short presentation of the scientific project (maximum four pages):

RBS analysis of doped ZnO thin films grown by pulsed electron beam deposition

In the last years we have studied the growth and physical properties of wide band gap oxide thin films for use in a wide range of applications in electronics, optoelectronics, spintronics, biomedical applications, nanotechnology, etc. [1-8]. These oxide thin films have been grown by pulsed electron beam deposition method (PED), a growth technique developed recently at INFLPR, which has features similar with the pulsed laser deposition (PLD) but uses a pulsed electron beam instead of a laser beam [1]. Our researches were focused on the use of the PED method for oxide thin films growth with a good control of the cationic composition, oxygen stoichiometry, surface morphology and crystalline structure of the films [1-8]. In particularly, we have shown by means of Rutherford backscattering spectrometry (RBS) performed at 2.5 MeV Van de Graaff accelerator of the SAFIR IBA Laboratory, University Pierre and Marie Curie, that the composition of the various oxide thin films grown by PED is close to that of the target, even in the case of complex oxide compounds [1-8]. Moreover, the growth of complex oxide thin films with different functional physical properties was performed, with special attention paid to the epitaxial growth of oxide thin films on single crystal substrates [2-4].

We have put in evidence a metal-semiconductor transitions (MST) in epitaxial un-doped ZnO and In_2O_3 thin films grown by PED on c-cut sapphire single crystal substrates and ZnO on MgO single crystal substrates, respectively [2-4].

As example, Figure 1 a represents a typical RBS spectrum recorded on a ZnO film grown by PED on a sapphire substrate at 300 °C [2], and the shape of this spectrum, i.e., the sharp front edge of the Al contribution and rear edge of the Zn peak, indicates the absence of any interdiffusion between film and substrate or large surface roughness. This spectrum shows the formation of a film with a uniform in-depth distribution of Zn and O species without measurable amounts of any other elements, i.e., the incorporation of impurities in the film during the PED process, which could play an important role in the transport properties of the films, is not evidenced in the RBS analyses.



(a) (b)

Figure 1. (a) A typical RBS spectrum (solid rectangles) recorded for a ZnO film grown by PED on a c-cut sapphire substrate at 300 °C [2] and the corresponding fit (solid line); (b) Temperature dependence of the resistivity ρ(T) of the same film. The solid line is the fit with quantum corrections to conductivity model of the experimental data (open rectangles) [2].

Moreover, the RBS spectrum is well fitted with the Zn_1O_1 composition, but due to the limited accuracy of RBS in the determination of the content of light elements such as oxygen (about 4%), a slight oxygen deficiency in the zinc oxide cannot be excluded [2].

The interpretation of this MST in the frame of the quantum corrections to conductivity in disordered oxides was addressed and correlated to the growth mechanism by PED [2, 4] (Figure 1b).

Largely oxygen deficient indium tin oxide thin films (more than 20% oxygen missing) grown by PED lead to the synthesis of nanocomposite films, i.e. metallic (In, Sn) clusters embedded in a stoichiometric and crystalline oxide matrix [5, 6]. The presence of these metallic clusters induces specific transport properties, i.e. a metallic conductivity via percolation with a superconducting transition at low temperature (about 6 K) and the melting and freezing of the In-Sn clusters in the room temperature to 450 K range evidenced by large changes in resistivity and a hysteresis cycle [5, 6].

Based on these approaches we intend to demonstrate that by a precise control of the nature and concentration of dopants, oxygen deficiency and structural defect density it is possible to tune the physical properties of doped ZnO thin films and to give them new functionalities for applications.

(i) Multifunctional ZnO thin films

Owing to its specific optical (high transparency in the visible domain) and electrical (high conductivity) properties, ZnO is used in a lot of applications in thin film form (transparent thin film transistors, diodes, sensors, etc.) [10]. Doping by well thought elements has been used to enhance the physical properties of ZnO or to induce a new one. Indeed, doping with Al, Ga, Si,...has been used to increase the conductivity of ZnO thin films; doping with Co, Mn, Ni, has induced ferromagnetism to obtain dilute magnetic semiconductors; while the doping with Er has been used in view of optoelectronic applications [10-12]. Our approach will be to use rare earth like Nd as doping element as it could lead to multiple applications:

- Nd^{3+} should be an electron donor substituted to Zn^{2+} in the ZnO matrix, to increase the electrical conductivity;
- Nd is a magnetic element in view of the formation of dilute magnetic semiconductor;
- Nd can be used as a photonic convertor for photovoltaic applications.

These three possible effects would lead to multifunctional ZnO thin films, and an important point will be the precise determination of the doping level and film composition in order to understand the pertinent factors and phenomena affecting the electronic, magnetic and optical properties of such doped ZnO films. These insights (doping concentration and profile, film composition) will be obtained by Rutherford Backscattering Spectrometry (RBS).

(ii) **Ion beam analyses**

The important point would be the precise determination of the cationic and oxygen composition of such films. Thus, the requirements for RBS analysis are: $^{4}He^{+}$, 1.8 MeV, about 30 nA on 1 mm^2 spot size, with the possibility to analyze 10-12 samples in a single batch. Such conditions will lead to a 4% accuracy on the determination of the oxygen composition in our films with a duration of RBS analysis of about 15-20 min per sample. This oxygen accuracy is sufficient for our studies on doped ZnO thin films which are focused on their physical properties [2].

Other requirements for experiments are: 17 keV resolution for the detector, possibility to measure the dead time for each RBS spectrum, RBS spectrum with 512 channels, spectrum data in format ".asc" for future treatment by a simulation program. The simulation of the RBS spectra will lead to the determination of these parameters with an accuracy of 1 and 4 % for the cationic and oxygen composition respectively.

We estimate at 8 hours (1 day) the RBS measurements at each two weeks interval in order to have a rapid feedback to the optimization of the doped ZnO thin films grown by PED method and correlation with other thin film investigations.

References:

[1] M.Nistor, N.B.Mandache and J. Perrière, Pulsed electron beam deposition of oxides thin films, J. Phys. D: Appl. Phys. **41** 165205 (2008)

[2] M. Nistor, F.Gherendi, N.B.Mandache, C. Hebert, J. Perrière, W.Seiler Metal-semiconductor transition in epitaxial ZnO thin films, J.Appl. Phys., **106**, 103710 (2009)

[3] W.Seiler, M.Nistor, C.Hebert, J.Perrière, Epitaxial undoped indium oxide thin films: Structural and physical properties, Solar Energy Materials&Solar Cells **116**, 34 (2013)

[4] M. Nistor, NB Mandache, J. Perrière, C. Hebert, F.Gherendi, W. Seiler, Growth, structural and electrical properties of polar ZnO thin films on MgO (100) substrates, Thin Solid Films, **519** 3959 (2011)

[5] M. Nistor J. Perrière, C. Hebert, W. Seiler, Nanocomposite indium–tin oxide films formation induced by a large oxygen deficiency, J.Phys.Cond. Matt. **22**, 045006 (2010)

[6] M. Nistor, A.Petitmangin, C. Hebert and W. Seiler, Nanocomposite oxide thin films grown by pulsed energy beam deposition, Appl.Surf.Sci. **257**, 5337 (2011)

[7] M. Nistor, F. Gherendi, N.B. Mandache, Angular distribution of species in pulsed energy beam deposition of oxide films, Appl. Surf. Sci. **258** 9274 (2012)

[8] M. Nistor, Doped zinc oxide thin films as transparent conductive electrodes, Rom.Rep.Phys. **64**, 1313 (2012)

[9] E.Millon, M. Nistor, C. Hebert, Y. Davila and J. Perriere, Phase separation in nanocomposite indium tin oxide thin films grown at room temperature: on the role of oxygen deficiency, J. Mater. Chem. **22**, 12179 (2012)

[10] Ü. Özgür, Ya. I. Alivov, C. Liu, A. Teke, M. A. Reshchikov, S. Doğan, V. Avrutin, S.-J. Cho, H. Morkoç, A comprehensive review of ZnO materials and devices, J. Appl. Phys. **98**, 041301 (2005)

[11] J. Clatot, G. Campet, A. Zeinert, C. Labrugère, M. Nistor, A. Rougier, Low temperature Si doped ZnO thin films for transparent conducting oxides, Solar Energy Materials and Solar Cells, **95** 2357 (2011)

[12] R. Perez-Casero, A. Gutierrez-Llorente, O. Pons-Y-Moll, W. Seiler, R.M. Defourneau, D.Defourneau, E. Millon, J. Perriere, P. Goldner, B. Viana, Er-doped ZnO thin films grown by pulsed-laser deposition, J. Appl. Phys. 97, 054905 (2005)

Beam time request (unit=8 hours)* 8h x2/month **Desired Period* 01/11/2013-01/06/2014**

Desired beam properties :

Type* 4He+ **Energy(MeV)*** **1.8 MeV** **Intensity*(p/nnA) 30 nnA on 1 mm^2 beam spot**

Vacuum Requests* 10^{-6} mbar

Special requirements for detectors, electronics, aquisition system: 17 keV resolution for the detector, possibility to measure the dead time for each RBS spectrum, RBS spectrum with 512 channels, spectrum data in format ".asc" for future treatment by a simulation program.

Minimal information needed for the radiological risk evaluation:

a) Source activity:*

b) Use of open sources* no

c) Estimate of the residual activity as a result of iradiation*

d) Means of storage/transportation for irradiated targets* no