BEAM REQUEST at Bucharest TANDEM 3MV

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

Experiment Responsable:

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

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

In the first session of experiments at Bucharest IFIN-HH TANDEM 3MV we have performed Rutherford Backscattering Spectrometry (RBS) measurements of doped or not doped ZnO thin films grown by pulsed electron beam deposition method (PED) under different growth conditions in order to get a rapid feedback for the optimization of the thin films and their correlation with other thin film investigations. PED is 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 previous researches demonstrated that the PED is a growth method allowing a very good control of the cationic composition, oxygen stoichiometry, surface morphology and crystalline structure of oxide thin films [1-6]. Moreover, a special attention was paid to the epitaxial growth of oxide thin films on single crystal substrates, obtaining different functional physical properties of films [2-5].

Owing to its specific optical (high transparency in the visible domain) and electrical (high conductivity) properties, zinc oxide (ZnO) is used in a lot of applications in thin film form (transparent thin film transistors, diodes, sensors, etc.) [7]. Doping by well thought elements has been used to enhance some physical properties of ZnO or to induce new ones. Actually, 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 [7-9]. Our approach was 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.

In order to check if these three possible effects would lead to multifunctional ZnO thin films, an important point was the precise determination of the doping level and film composition for understanding the pertinent factors and phenomena affecting the electronic, magnetic and optical properties of such doped ZnO films.

The doping concentration, and depth profile of the various elements were obtained by RBS in following experimental conditions: $^{4}He^{++}$, 1.8 MeV, about 30 – 90 nA on 0.5 - 1 mm^2 spot size. The weak points of the first RBS measurements: (a) long time for stabilization of the source at required parameters and therefore the late starting of experiments and (b) variation of the current during recording a RBS spectrum will be certainly improved in next experiments. The thin films were analyzed in multiple batches because the sample support supports the mounting of maximum 4 samples/measurement, depending on the size of the sample. The simulation of the RBS spectra was done by using the RUMP software.

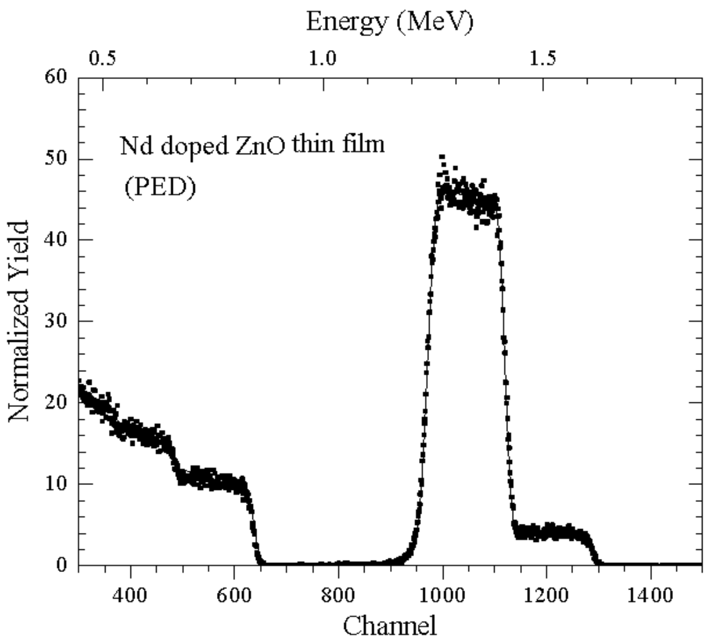

Figure 1. A typical RBS spectrum (line) recorded for a Nd doped ZnO film grown by PED on a Al_2O_3 single crystal substrate at room temperature and the corresponding fit (solid rectangles);

Figure 1 shows a typical RBS spectrum recorded on a Nd doped ZnO thin film grown by PED on a Al_2O_3 single crystal substrate at room temperature and under 10^{-2} mbar oxygen. The shape of this spectrum indicates the presence of a slight surface roughness of the film, which was included in the fit model. Moreover, this spectrum shows the formation of a film with a uniform in-depth distribution of Zn, Nd and O species ($Zn_{0.978}Nd_{0.022}O_{1.000}$ composition), which could explain the high sheet resistance measured for this film. Actually, a stoichiometric film is grown in these conditions, and the carrier density is low as a noticeable concentration of oxygen vacancies (electron donors) is not present. Amounts of any other elements, i.e. incorporation of impurities in the film during the PED process which could deteriorate the film properties, were not evidenced in the RBS spectrum.

A second approach was based on the work presented in a previous paper, where we have shown that the thin film thicknesses profile of PED, obtained by RBS measurements, has a slightly broader shape than that obtained in the case of PLD [6]. Because the preservation of the same composition all along the major axis of the ZnO films is required in applications, films were scanned along their major axis at 2 – 4 mm distance between two ion irradiations, and for each point a RBS spectrum was recorded. Thus, film thickness and distribution of the various elements as a function of depth were obtained by simulation of the RBS spectra.

A typical example of film thickness profile as a function of position on the substrate is presented in Figure 2 for a Nd doped ZnO thin film grown by PED on a Si substrate at room temperature.

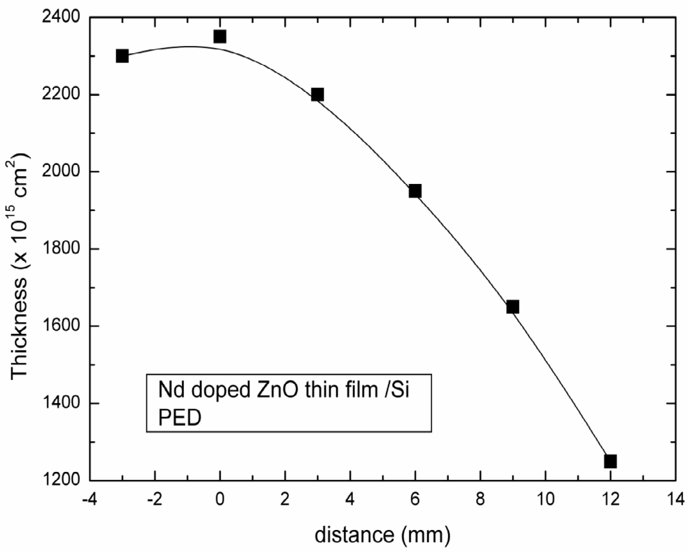

Figure 2 Thin film thicknesses profile for a Nd doped ZnO film grown by PED on a Si substrate at room temperature (the line is a guide for eye)

These results show the importance of rapid feedback of the RBS analysis to the optimization of the thin films and their correlation with other thin film investigations (electrical, optical, magnetic properties, etc.).

For the next RBS experiments we intend:

(i) to continue the analysis of Nd doped ZnO thin films grown by PED at intermediate substrate temperatures, which were not analysed in the first RBS session. These measurements will be very useful to clarify the limits of the Nd doping in ZnO thin films;

(ii) to test other rare earth elements doped ZnO thin films and to correlate them with electrical and optical properties;

(iii) to test the doping of ZnO thin films with Fe or Co elements in the view of inducing ferromagnetism and to correlate the composition with the structural and physical properties.

Ion beam analyses

The aim of the proposal is the precise determination of the cationic and oxygen composition of doped oxide thin films. The requirements for RBS analysis are: $^{4}He^{++}$, 1.8 - 2 MeV, about 50 nA on 1 mm^2 spot size, with the possibility to analyze the samples in multiple measurements (batches), taking into account that the sample support allows up to 4 samples/measurement. Such conditions will lead to a 4% accuracy on the determination of the oxygen composition in our films, which is sufficient for our studies focused on the physical properties of doped oxide thin films.

Other requirements for experiments are: 17 keV resolution for the detector, possibility to measure the dead time for each RBS spectrum, RBS spectrum with 1024 channels.

We estimate for the RBS measurements 8 hours (1 – 2 days, depending on the time necessary for the source stabilization) in each month to have a rapid feedback to the optimization of the doped oxide 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, F. Gherendi, N.B. Mandache, Angular distribution of species in pulsed energy beam deposition of oxide films, Appl. Surf. Sci. **258** 9274 (2012)
[7] 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)
[8] 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)
[9] 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 x1/month **Desired Period* 01/09/2014-31/12/2014**

Desired beam properties :

Type* 4He++ **Energy(MeV)*** **1.8 MeV – 2 MeV** **Intensity*(p/nnA)** 50 nA **on 1 mm^2 beam spot**

Vacuum Requests* 10^{-6} mbar

Special requirements for detectors, electronics, acquisition system: 17 keV resolution for the detector, possibility to measure the dead time for each RBS spectrum, RBS spectrum with 1024 channels

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