 ROBL-CRG	Experiment title: Real-time evolution of ZnO:Al thin film structure and electrical properties during annealing	Experiment number: 20_02_651
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Report

Application of transparent conductive oxides in advanced photovoltaic cells and flat-panel displays requires thin films with high transmittance (>90%) and low resistivity ($\sim 10^{-4}$ Ohm-cm). Reorganization and improvement of the film structure should be key factors for enhancement of the electron mobility [1], however it requires additional investigation. The aim of the experiment is relation of the ZnO:Al film structure (texture, grain size etc.) with the film electrical resistivity. For this propose the changes of structure and electrical resistivity of ZnO films with different initial crystalline quality and Al content were studied during annealing in vacuum using *in-situ* techniques.

Undoped ZnO films were deposited by medium frequency pulsed reactive magnetron sputtering at the base pressure of 3.4×10^{-7} mbar using a mixture of Ar and O₂ (partial pressure of 7.4×10^{-3} mbar and 1.4×10^{-3} mbar, correspondingly). The substrates were 10 mm x 10 mm x 0.3 mm pieces of Si(100) single crystals covered by 200 nm of thermally grown SiO₂ layer. The ZnO films were implanted with aluminium ions at ion energy of 110 keV and fluences of 1×10^{16} and 5×10^{16} Al⁺ ions/cm².

The implanted samples were characterized by *in-situ* synchrotron XRD analysis (photon energy of 8.045 keV) and simultaneous four-point probe measurements during non-isothermal (T-ramp: 13 K/min) and isothermal (temperature range of 450–800°C) annealing.

The x-ray diffraction patterns of as-implanted ZnO films demonstrate only a (002) ZnO peak with FWHM in the range of $0.3 - 0.55^\circ$ depending on the deposition temperature as well as implantation fluence. Figures 1 demonstrates the intensity and FWHM of ZnO (002) peak as a function of annealing temperature. The XRD patterns show no structural changes below the temperature of 450°C . A further increase of annealing temperature leads to growth of the (002) peak intensity and decrease of FWHM. However, the recrystallization rate of the low-fluence implanted films ($1 \times 10^{16} \text{ Al}^+/\text{cm}^2$) is essentially higher than that of the samples implanted at $5 \times 10^{16} \text{ Al}^+/\text{cm}^2$. The low-fluence implanted ZnO films become structurally identical after annealing at maximum temperature of 880°C and there is no difference between the films deposited at 250 and 550°C . The four-point probe measurements (not shown here) demonstrate significant decrease of the film resistivity with increasing temperature at the very early stage of non-isothermal annealing ($100 - 300^\circ\text{C}$) before an outset of recrystallization. Further increase of temperature leads to the crystallinity improvement at relatively weak resistivity changes.

The evaluation of the XRD and four-point probe data and calculation of the kinetic parameters of recrystallization as well as the activation energy are in progress.

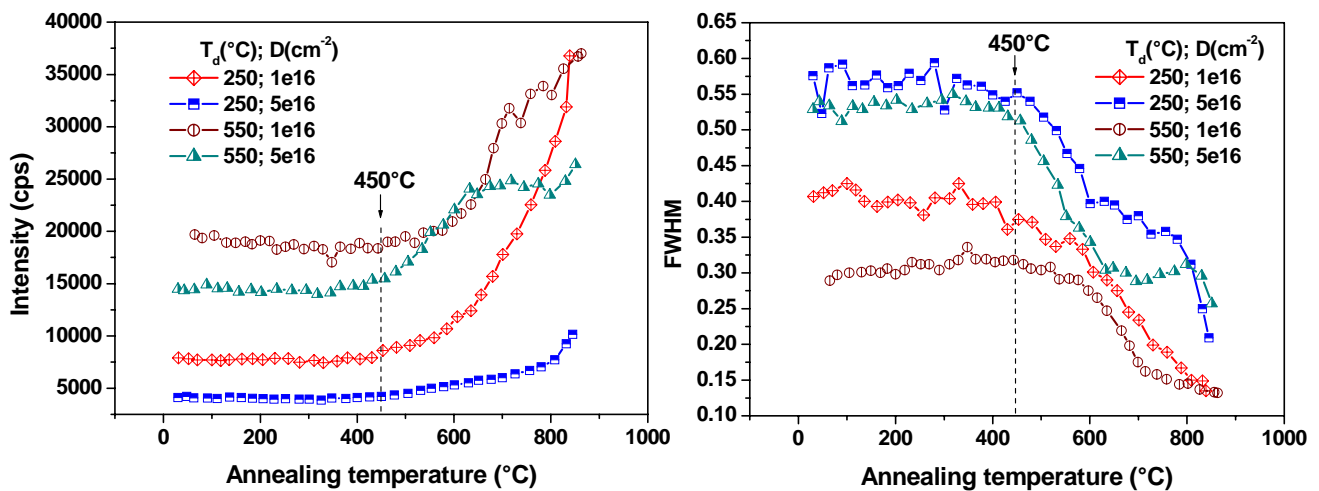


Fig. 1: The temperature dependence of the XRD intensity and FWHM of the ZnO (002) peak of Al^+ -implanted samples during non-isothermal annealing (T -rate: 13 K/min).

References

1. Birkholz M. et al, Phys. Rev. B 68, 205414 (2003)