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Microstructure and strain relaxation in thin nanocrystalline platinum films produced via different sputtering techniques

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In this study we investigated the correlation between microstructure and residual strain relaxation in nanocrystalline Pt films with a thickness of about 20 nm produced by different deposition techniques: magnetron sputtering and ion beam sputtering. X-ray diffractometry was carried out using synchrotron radiation. The out-of-plane interplanar distance was measured during isothermal in situ annealing at temperatures between 130 °C und 210 °C. The thermoelastic expansion coefficient is equal for both types of nanocrystalline Pt films and slightly lower than for coarse grained Pt. The relaxation of residual out-of-plain strain depends on temperature and is significantly stronger in the case of the magnetron sputtered films than for the ion beam sputtered films. Different relaxation of compressive stress is ascribed to the different microstructures which evolve during deposition via the corresponding deposition technique. Thickness fringes around the (1 1 1) Bragg peak deposited via magnetron sputtering reveal that these films are essentially composed of columnar (1 1 1) oriented grains which cover the whole film thickness. In contrast, no thickness fringes are observed around the (1 1 1) Bragg peak of films prepared by ion beam sputtering indicating a significantly different microstructure. This is confirmed by Electron Backscatter Diffraction which reveals a (1 1 1) texture for both types of films. The (1 1 1) texture, however, is significantly stronger in the case of the magnetron sputtered films. Grain growth at low homologous temperatures is considered to be an important contribution to relaxation of residual stress.

1. Introduction

In the present work we investigate residual strain relaxation of thin Pt films deposited by different deposition techniques: magnetron sputtering and ion beam sputtering. Residual stress and strain are typical characteristics of film substrate combinations. The origin of residual stress in thin metal films is widely discussed in literature [1–7]. Metal films are commonly assumed to develop via Vollmer–Weber growth mode [7]. At the beginning of deposition discrete islands of the metal are built on the substrate. With increasing deposition time the islands grow. Growth of the islands adhered to the substrate is associated with stress which is commonly compressive [4–6]. After impingement, coalescence of the islands takes place: the island built a grain boundary to reduce the surface. Coalescence is associated with tensile stress [8]. Further deposition leads to a continuous film. The residual stress of the final film can be either tensile or compressive and depends on the deposition conditions [9]. On one hand, post-annealing of the metal films adhered to the substrate induces thermal stress. On the other hand, relaxation processes of the intrinsic (growth related) stresses can take place since thermal energy is available.

In the present work, we deposited Pt films with a final thickness of about 20 nm using magnetron sputtering or ion beam sputtering. The residual stress of the as-deposited films is compressive for both deposition techniques. After deposition we performed X-ray diffraction measurements (XRD) during in situ annealing. The out-of-plane interplanar distance was determined for the Pt films as-deposited, during in situ annealing and after cooling down to room temperature. From the diffractograms information on the microstructure and the change of the microstructure during

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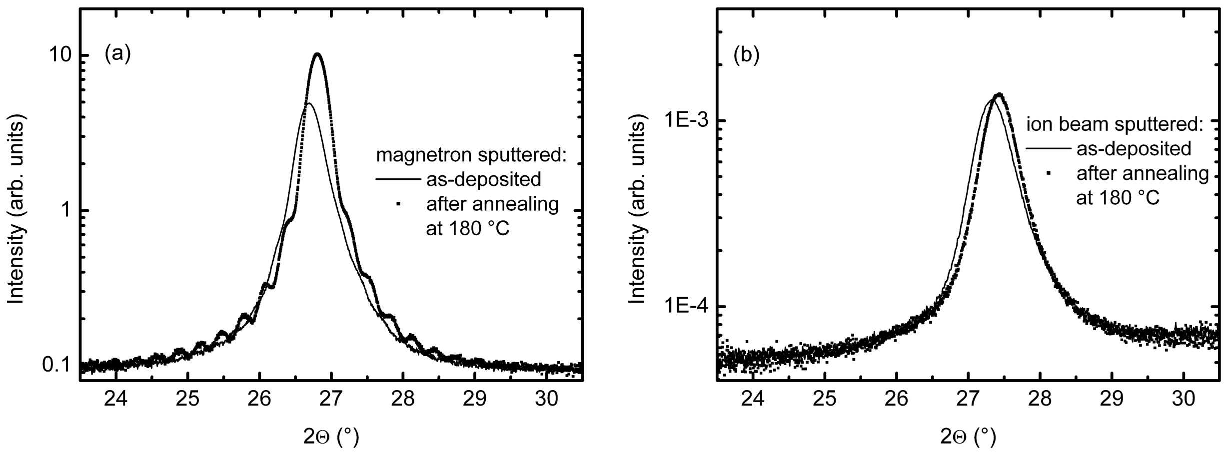

Fig. 1. (1 1 1) Bragg peak of a 20 nm Pt film as-deposited (solid line) and after annealing at 180 °C for 10 h and cooling down (dotted line) deposited via magnetron sputtering (a) and ion beam sputtering (b).

in situ annealing is deduced. The amount of residual strain which is released during in situ annealing at a given temperature is significantly different for the two types of Pt films. Release of residual strain is related to the microstructure.

2. Experimental

Ion beam sputtered Pt films with were deposited in argon using a commercial ion beam coater (Gatan IBC 651) at 5 keV and a working pressure of 0.5 Pa (base pressure <5×10^{-4} Pa). The deposition rate was 0.7 nm/min and the thickness of the films was about 20 nm. Magnetron sputtered films were prepared at the Paul Scherrer Institute (PSI), Switzerland, using dc magnetron sputtering. Here, sputtering was carried out in an argon atmosphere at a power of 50 W and a working pressure of 0.3 Pa (base pressure <4×10^{-4} Pa). The deposition rate was 0.5 nm/s and the thickness of the films was about 20 nm. Commercially available oxidized silicon wafers were used as substrates in order to impede silicide formation during post-annealing. During deposition the substrates were at room temperature. The thickness of the films was measured using X-ray reflectometry.

XRD experiments in $\Theta/2\Theta$ geometry were carried out at the Rossendorf beam line at ESRF (BM20), Grenoble, using a six-circle goniometer (HUBER) and a parallel beam. The X-ray energy was 11,500 eV corresponding to a wave length of 1.08 Å. The samples were placed on a heater. Sample temperature was measured using a type K thermocouple mounted at the sample surface. Temperature was controlled using a Eurotherm 2804 temperature controller. Sample and heater were inside a Kapton dome which was evacuated. During heating a vacuum of about 4×10^{-4} Pa was achieved. Rocking curves and *z*-scans were measured to align the sample. Afterwards XRD was measured for the as-deposited sample. After heating up the sample was aligned again and XRD was measured. During in situ annealing alignment and XRD measurement were alternately repeated. After cooling down to room temperature the sample was aligned again and XRD was measured. The successive alignment was indispensable in order to obtain reliable measurements. Since the beam current at the synchrotron varies with time the intensity of the primary X-ray beam is monitored at the beamline. In order to compare the intensities of the Bragg peaks measured during a beam time the diffractograms were normalized to the monitor count rate. During beam time the settings of the monitor are unchanged. However, the intensities of Bragg peaks measured at different beam times cannot be compared directly due to different operation modes.

Electron Backscatter Diffraction (EBSD) measurements were carried out using a Nordlys EBSD detector of Oxford Instruments installed in a FEI Helios NanoLab 600. For these measurements four samples were investigated at a working distance of 10 mm and a specimen tilt of 71° at an acceleration voltage of 20 kV and a beam current of 1.4 nA. The EBSD system was calibrated by means of a single crystalline silicon sample. The data were recorded and processed using the CHANNEL5 suite of Oxford instruments. Inverse pole figures were calculated for each sample based on EBSD mappings of 10,000 points.

3. Results

As mentioned, XRD was measured in $\Theta/2\Theta$ geometry. In this geometry only atomic planes parallel to the sample surface contribute to the diffractograms. This means that the out-of-plane interplanar distance is probed. We measured the (1 1 1) Bragg peak which is by far the strongest peak in the diffractograms. Fig. 1a shows Bragg peaks of a Pt film with a thickness of about 20 nm deposited via magnetron sputtering. The solid line is the Bragg peak of the Pt film as-deposited and the dotted line is the Bragg peak of the Pt film after annealing at 180 °C for 10 h and cooling down to room temperature. The intensity of the Bragg peak is significantly increased after annealing (note the logarithmic scale). The peak position is shifted to a higher 2Θ value. This means that the out-of-plane interplanar distance is decreased. The Bragg peaks are asymmetric and show characteristic interference fringes. These thickness fringes arise if a film is composed of columnar grains with uniform size and orientation. The fringes are already present in the Bragg peak of the as-deposited film and are much more pronounced after annealing. To determine the out-of-plan interplanar distance we fitted the peaks using a split pseudo-Voigt function and calculated it from the peak position using Bragg's law. Fig. 1b shows the corresponding Bragg peaks of a 20 nm Pt film deposited via ion beam sputtering in the as-deposited state (solid line) and after annealing at 180 °C for 10 h and cooling down to room temperature (dotted line). Since the measurements for the two types of films were performed at different beam times the intensities cannot be compared directly and are given in arbitrary units (see Section 2). In contrast to the magnetron sputtered film, no fringes can be observed at all around the Bragg peaks of the ion beam sputtered film. The intensity of the Bragg peak is only slightly increased but the peak position is also shifted to a higher 2Θ value. This shift, however, is significantly smaller than in the case of the magnetron sputtered Pt film.