NEW MAGNETRON CONFIGURATIONS FOR SPUTTERED Nb ONTO Cu

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Abstract

Niobium sputtered film microstructure and morphology and consequently its superconducting properties, strongly depend on target-substrate deposition angle.

In order to improve the Nb film quality for 1,5GHz cavity coatings, we investigated the application of three main ideas to the sputtering process: i) making niobium atoms impinging perpendicularly the substrate surface, ii) promoting the effect of plasma bombardment of the growing film, iii) increasing the sputtering rate.

Therefore, several different sputtering configurations are under development.

The effect of Nb atoms arriving perpendicular to the substrate is explored either by using a cathode that follows the cavity shape or by increasing the plasma confinement efficiency by means of a target parallel to the magnetic field lines.

The removal of adsorbed impurities from the film surface and the increase of the film density are investigated by a biased third electrode that promotes the positive ions bombardment of the growing film. A mixed bias-magnetron has been built using a positively charged metal grid positioned all around the cathode.

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keywords: niobium superconductor, Nb/Cu cavities, magnetron, bias, sputtering, pulsed current mode

INTRODUCTION

The adoption of the Nb/Cu technology at CERN for the LEP cavities and the successful operation of ALPI Linac at INFN-LNL have demonstrated the feasibility of using large-scale copper accelerating cavities coated with a thin superconducting niobium film. At low fields, the BCS surface resistance of Nb/Cu cavities is lower than for bulk niobium cavities at the same RF frequency. Due to the small grain size, high process gas content and some oxygen contamination of the films, the mean free path of the sputtered Nb layer is lowered to 25–50 nm, corresponding to a *Residual Resistivity Ratio (RRR)* of 10–20 [1]. This is the range where the BCS surface resistance reaches a minimum. The Nb/Cu films drawback - commonly referred to as the Q-slope problem - is that the surface resistance depends on the RF magnetic field[2].

One limit of the standard cylindrical magnetron sputtering deposition technique is the coating geometry, in fact assuming a cosine law for the atom emission mechanism, the incidence angle of the atoms on the cavity wall varies from 9±4° around the equator to 50±10° near the iris.

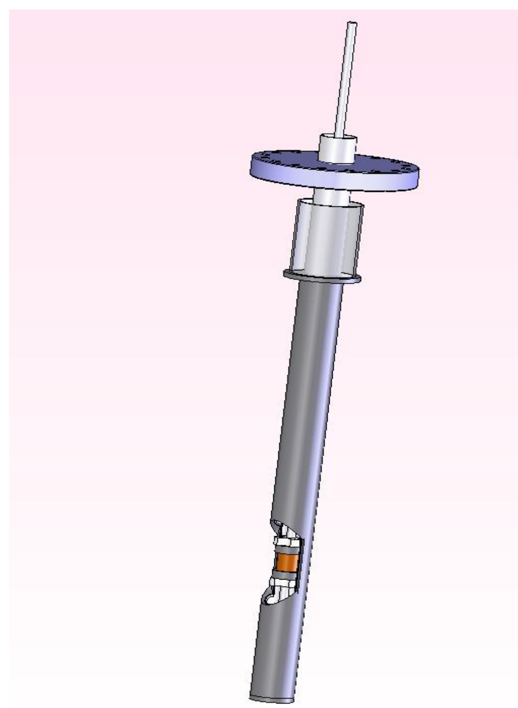

Figure 1: Standard CERN cylindrical magnetron configuration. The cathode section shows the NdFeB magnet inside the niobium tube.

In a previous study a decrease in superconducting properties of niobium films has been observed when increasing the deposition angle between target and substrate, this effect is clearly due to change in the coating morphology and reproduce the film behaviour in cavity cell [3]. This dependence is confirmed by X-ray diffractometry, AFM analysis, magneto-optical images and electrochemical impedance spectroscopy.

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Moreover texture analysis show clearly the sputtered film directional growing: sputtered films tend to grow with the normal to the 110 crystal plane aligned to atoms arrival direction [4]. On the contrary in arc deposition the substrate is negatively biased so the electric field lines intersect the surface of substrate holder always at right angles; as a consequence ions collide nearly perpendicularly to the substrate regardless of the angle between target and substrate and preferential orientation shows no angular dependence.

To explain theoretically the thin films behaviour ,the key parameters are the electrons mean free path l in normal state and the coherence length ξ. Low values of l/ξ give high Q and higher slope. For film coated cavities there is no hope to get rid of the slope, unless RRR is increased, but in this case Q values will be lower than the actual value [5].

The CERN standard film coating procedure consists of covering the inner wall of copper resonator using the cylindrical magnetron sputtering technique [6]. The aim of the present work is to improve the film quality modifying or completely changing the coating configuration starting from three main ideas:

- making niobium atoms impinging perpendicularly the substrate surface;
- promoting the effect of plasma bombardment of the growing film in order to remove impurities weakly bonded to the surface;
- increasing the sputtering area and the plasma ionization efficiency in order to increase the sputtering rate.

DEPOSITION TECHNIQUE: MAGNETRON SPUTTERING

Since film morphology is strictly correlated to the deposition angle and electrical and superconducting film properties degrade vs. deposition angle, comprehension of sputtering principles is compulsory for conceiving new magnetron configurations.

Sputtering Technique

In sputtering deposition technique magnets are added to enhance the plasma ionization efficiency by increasing the electron path lenght: an electron in a uniform magnetic field B will drift along the field lines with a speed $v_{||}$ that is unaffected by the field and orbit them at a gyro or Larmour radius ($r_g = 3.37\text{x}10^{-6}\ W_\perp^{1/2}/B$), and with a gyro or cyclotron angular frequency (ω_c=1.76x10^{11}B), where $W_\perp$ is the energy associated with the electron motion perpendicular to the field.

Near the target surface electrons are reflected by both electric and magnetic fields. Electrons tend to conserve their magnetic moment μ_M ($\mu_M = m_e v_\perp^2/B$); thus conservation of energy may cause electrons passing in the direction of $\nabla B_{||}$ to be reflected by magnetic field before they reach the cathode surface.

When there is a component of electric field $E_\perp$ perpendicular to B, the drift of speed $v_E = E_\perp/B$ develops in a direction perpendicular to both $E_\perp$ and B and combines with the orbiting motion. This is the $(E\text{x}B)/B^2$ drift collective motion.

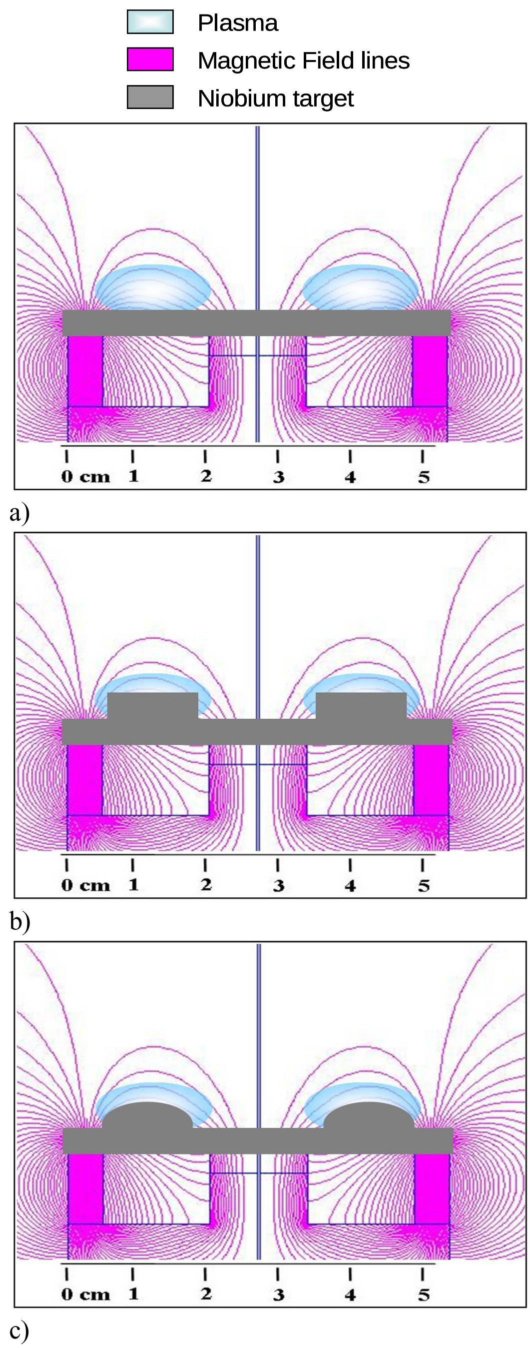

Figure 2: Magnetic field lines simulations for the 2 inches planar magnets with three different shapes (Pandira Software[7]). 2a): Planar target. 2b): Squared target. 2c): Rounded target

Since ions have much higher mass than electrons, only the latter are affected by magnetic fields used in magnetron devices (order of 100G). Over the range of parameter investigated, the current density at the cathode of the planar magnetron is peaked where the ExB drift is maximum. Accordingly the higher sputtering rate is achievable where the magnetic field lines are tangent to the surface, as well as perpendicular to the electric field.[8,9]

Sputtering rate

The fraction f_i of impurities of species i trapped in a film is given by

$$f_i = \frac{a_i N_i}{a_i N_i + R} \qquad \textbf{Eq. 1}$$

where N_i is the number of atoms of species i bombarding unit area of film in unit time during deposition, α_i is the effective sticking coefficient of the species i during deposition and R is the deposition rate of the film. It is clear from Eq.1 that f_i can be reduced by increasing R [10].

According to this basic principle it's possible to test some magnetron configurations starting from 2 inches circular planar magnetron. The idea is to modify the target shape as it follows the magnetic field lines. In this way the electric field is always perpendicular to the magnetic field.

Three different target shapes has been tested: planar, squared and rounded (Figure 3). The three I-V curves have been compared.

Tests were performed in a cylindrical stainless steel vacuum chamber equipped with a planar magnetron source with a 2 inches wide niobium target. Argon has been used as process gas.

Magnetron I-V curves have been collected for the three different target shapes. Data fitted with the following power law[11,12] describe the current dependence with the applied voltage:

$$I = aV^b \qquad \textbf{Eq. 2}$$

where b is the index of performance of the electron trap and is as higher as the magnetic field confinement efficiency.

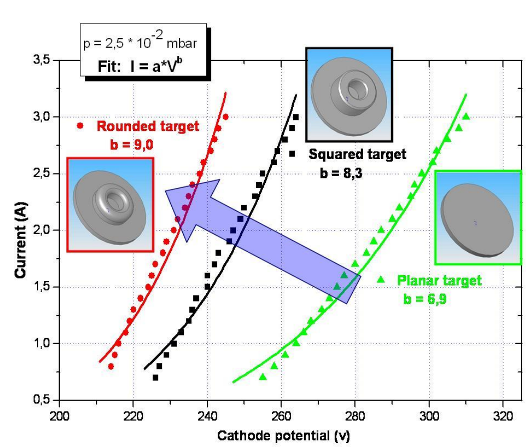

Figure 3: I-V curve of three different targets.

The curves showed in Figure 3 reveal information about the plasma ionization efficiency: the more efficient is the ionization the less is the voltage needed to reach a given cathodic current. That means that less power is needed to obtain a fixed deposition rate. The rounded target shows the best results

NEW MAGNETRON CONFIGURATIONS

Results obtained for 2 inches planar magnetron has been transposed to cylindrical magnetron for 1,5 GHz cavity coatings.

Original Apparatus

A stainless steel cavity-shaped deposition chamber has been mounted on a vacuum system and quartz substrates are positioned along two cavity shaped sample holders. An ultimate pressure of the order of 10^{-9} mbar is obtained after 30h bake-out at 150°C.

The cathode is located on the axis of the system. It consists of a vacuum tight stainless steel tube surrounded by a high purity niobium tube. The magnetron cathode is cooled by compressed air.

In the Cylindrical Magnetron (CM) configuration the extension tubes are coated first in an argon atmosphere of 1×10^{-2} mbar; a stable current of 1A is set between the cathode and the grounded sample holders. The magnet is moved in 14 steps over the cathode length in order to coat each tube uniformly. The cell is coated with slightly different discharge parameters, leaving the magnet fixed in its centre for 15 minutes: the argon pressure is reduced to 1.5×10^{-3} mbar so as to increase the mean free path of the atoms ejected from the cathode and the current is increased to 3A (400V) to rise the sputtering rate.

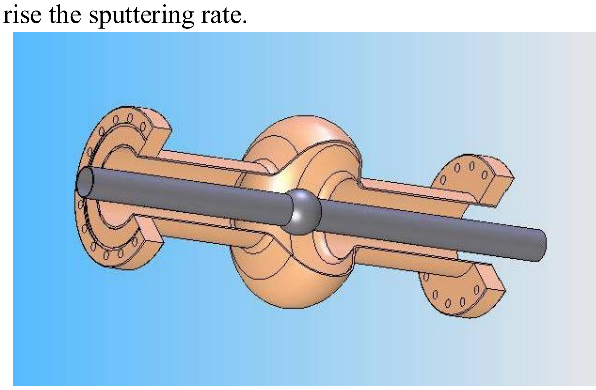

Figure 4: Schematic view of the cathode with the niobium ring.

Cavity shaped magnetron sputtering cathode

A niobium ring, 48.5 mm maximum diameter and 27 mm high, has been positioned around the cathode tube in the middle of the cavity cell as in Figure 4. The magnetic field is produced by a 2 cm high NdFeB permanent magnet located in the middle of the cell, inside the cathode, centred with the niobium ring

This configuration aims to reduce the deposition angle and to increase the area where the magnetic field is perpendicular to the electric field.

The glow discharge is established in argon atmosphere at a pressure of $1.5x10^{-3}$ mbar. A stable current of 3A is set between the cathode and the grounded cavity wall, corresponding to a voltage of 350 V.

Plasma is kept on for 5 minutes and then switched off for 10 minutes three times because of the high ring heating.

Film thickness measurement analysis show that, this configuration doesn't work as expected. The sputtering rate at the equator is higher than standard MC sputtering rate but, films are thicker in the equator than in the iris so the deposition is not homogeneous all over the ring.

Typical RRR and Tc results at the equator are 13 and 9.36K. Tc higher than Nb bulk means a compressive stress of the crystal lattice, confirmed also by X-ray diffraction cell parameter measure a = 3.29 Å respect to the bulk value a_0=3.303 Å. In addition texture analysis is performed on (110) peak, the most intense in Nb powder diffractogram, in order to investigate the occurrence of preferential orientation during film growth. Texture polar figures show the inhomogeneous grain orientation growth for all the positions along the cell.

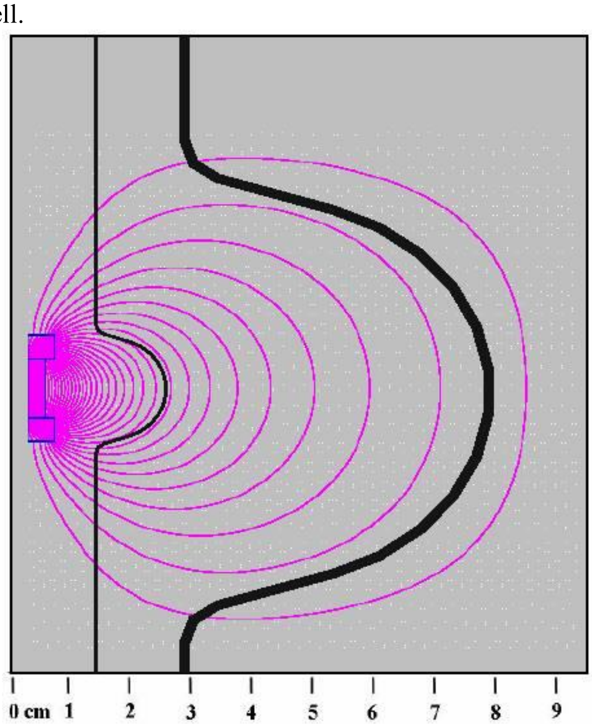

Figure 5: Magnetic field lines simulations for the 2cm high NdFeB magnet (Pandira Software[7]).

Several additional simulations have been run to optimize the best combination of magnet configuration and cathode shape.

Biased Magnetron Sputtering Configuration

If the film is given a negative potential with respect to the plasma, the resulting technique is referred to as bias sputtering. In this way films are subjected to a certain amount of resputtering. Advantages of bias are mainly the removal of most impurities during resputtering and rearrangement of atoms during film growing.

Considering the bias process Eq.1 is modified as

$$f_i = \frac{(a_i N_i - b)}{(a_i N_i - b) + R} \qquad \textbf{Eq. 3}$$

where b is a function of the bias current due to impurities ions [10]. The bias technique is highly reliable in fact over 40 Quarter Wave Resonators are installed and working at LNL for ALPI.

In our case the bias technique is added to the magnetron cathode as showed in Figure 66.

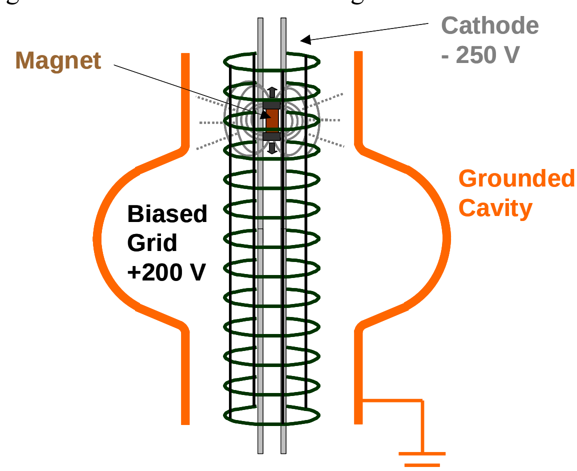

Figure 6: Scheme of the magnetron bias configuration.

Three grid configurations have been studied: stainless steel grid, titanium grid and stainless steel rods.

A stainless steel net has been rolled up and then welded on the end-tips. Despite the thick mesh the grid undergoes serious deformation during sputtering due to the high temperature reached by the cathode.

A titanium flat rhombus meshed net has been rolled up with an external diameter of 55 mm. This grid has also problem of deformation and, in addition, the mesh is even thicker that the first one causing shadowing during niobium sputtering.

The third and definitive more efficient grid configuration uses stainless steel rods: the niobium tube is surrounded by six vertical stainless steel rods isolated from the cathode by ceramic cylinders (Figure 7). Twelve springs keep the rods tight and avoid heating deformations. Tests with rods positively biased from 100V to 200V have been executed.

Eventual off-axis magnet magnetization could concentrate the electrons on one side of the cathode making only one of the rods being bombarded by all of them. The melting of the rods is avoided by rotating the magnet around its axis.

Depositions have been done in DC current mode and pulsed current mode. In the first mode a current of 2 A has been applied corresponding to a voltage of 360 V at the argon pressure of $4x10^{-3}$ mbar. With a bias voltage applied of 200V the electron current flowing on the rods is 14% of the cathode current.

Figure 7: Stainless Steel rectangular meshed grid, Titanium rhombus meshed grid, Stainless Steel rods with springs to keep them uptight.

Rising the bias voltage from 100V to 200V the electron current on the grid doesn't show relevant changes, meaning that it is on the asymptotic part of the I-V curve.

Pulsing the cathode with a 50 kHz square wave pulsed voltage is a way to increase the plasma ionisation degree and to promote the surface ionic bombardment during the positive voltage period.

RRR of 11 in the cell and 20 above and below the iris have been obtained after pulsed sputtering with the SS rods configuration. Maximum Tc obtained at the equator is 9.3K, meaning a compressive stress of the crystal lattice, comfiremd also by this X-ray diffraction cell parameter measure a = 3.30 Å. This results show no improvement or worsening of the film stress respect to MC standard deposition.

Texture analysis is performed on (110) peak, the most intense in Nb powder diffractogram, in order to investigate the occurrence of preferential orientation during film growth. Texture figures show an homogeneous grain orientation growth, mainly perpendicular to the substrate, all along the cavity cell.

Film thickness measurement show that the grid shadowing doesn't affect the sputtering rate in any position all along and around the cell.

Probable limiting factors are the impurities coming from the all the Stainless Steel immersed in the plasma; in fact they could partly mask the benefit effects of the applied bias. Impurity content analysis hasn't been executed yet.

Large Area Cavity Shaped Cathode

Test with different magnetron sputtering configuration allowed the project of a new sputtering configuration that combine all the tested ideas:

- promoting the growing film resputtering with a positively biased electrode
- granting the parallelism between cathode and substrate with cavity shaped surface cathode
- increasing the high sputtering rate with a Large cathode area

Bias sputtering of 1,5 GHz cavities was developed at CERN around 1985, it consisted on a rotating cavity shaped cathode. Its low ratio cathode/substrate area determines a very low sputtering rate (1 µm /day). One way to improve the film quality is increasing the sputtering rate by enlarging the sputtering area respect to the cathode one.

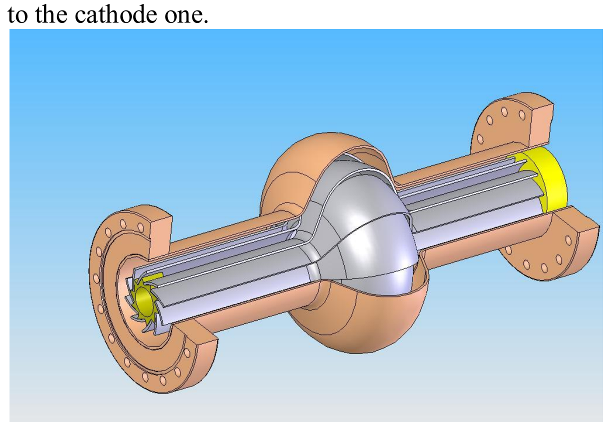

Figure 8: Schematic view of the cavity shaped cathode.

The idea under development is showed in Figure 8 and it consists of ten cavity shaped cloves cathode coaxial with the cavity. A stainless steel tube, coaxial with the cavity as well, is biased positively and it works as ion attractor and as support for the whole cathode system. The work is in progress.

CONCLUSIONS

Several approaches has been studied to improve the sputtered niobium film properties:

- making niobium atoms impinging perpendicularly the substrate surface;
- promoting the effect of plasma bombardment of the growing film in order to remove impurities weakly bonded to the surface;
- increasing the sputtering area and the plasma ionization efficiency in order to increase the sputtering rate.

Different magnetron sputtering configurations has been built and tested:

- Cavity shaped magnetron sputtering cathode
- Biased Magnetron Sputtering Configuration
- Large Area Cavity Shaped Cathode

Best results have been obtained with pulsed biased magnetron sputtering with the six rods configuration but improvement of the film purity is compulsory for emphasized the benefit of the ion bombardment.

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REFERENCES

[1] P. Darriulat 1998 Proc. 1998 European Particle Accelerator Conf. (Stockholm, Sweden) ed S. Myers et al.

[2] H. Padamsee, Supercond. Sci. Technol. 14 (2001) R28-R51

[3] D. Tonini, C. Greggio, G. Keppel, F. Laviano, M. Musiani, G. Torzo, V. Palmieri; "*Morphology of Niobium Films Sputtered at Different Target – Substrate Angle*" Proceeding of 11th Workshop on RF Superconductivity, Lubek (Germany) ; Diploma thesis "*Sputtered Niobium Film Morphology for Different Target-Substrate Deposition Angles*" Diego Tonini, LNL-INFN, Facoltà di Scienza dei Materiali, Università degli Studi di Padova.

[4] G. Keppel, M. Musiani, N. Patron, V. Palmieri, D. Tonini, G. Torzo, "*Comparison Between Morphology of Superconducting Niobium Films Deposited by Magnetron Sputtering and Cathodic Vacuum Arc*"; Proceedings of XVII AIV congress, Venezia Lido, 28 June- 2 July 2004.

[5] V. Palmieri, Proceeding of 12th Workshop on RF Superconductivity 2005, Ithaca (New York, USA)

[6] C. Benvenuti, S. Calatroni, P. Darriulat, M.A. Peck, A-M. Valente, C.A. Van't Hoff "*Study of Residual Surface Resistance of Niobium Films at 1.5 GHz"*, Physica C 351 (2001) 421-428

[7] James H. Billen and Lloyd M. Young; Poisson Superfish – Pandira; LA-UR-96-1834 Revised November 5, 2003; Los Alamos National Laboratory

[8] Penfold A. S 1995 *Handbook of Thin Film Process Technology* (Bristol: IOP Publishing) A3.2.1

[9] A.E.Wendt, M.A. Lieberman and H. Meuth, J. Vac. Sci. Technol. A6, 1827 (1988)

[10] L. I. Maissel and R. Glang, Handbook of Thin Film Technology, McGraw-Hill, New York, 1970.

[11] John A. Thornton, "*Coating Deposition by Sputtering*", Hanbook of plasma processing technology, Stephen M. Rossagel, Jerome J. Cuomo and William D. Westwood eds, Noyes Publications, 196, (1990)

[12] S. M. Rossnagel and H.R.Kaufman, J. Vac. Sci. Technol. A 6(2) 223 (1988)