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EFFECT OF DEPOSITION RATE ON THE STRUCTURE AND RESISTIVITY OF SPUTTERED Ti FILMS

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The new generation of low pressure and high–deposition rate magnetrons make it possible to investigate the effect of decreasing the operating pressure and the deposition rate (a_D) on the properties of sputtered films. This knowledge is of principal importance for the industrial scaling of the sputtering process, i.e. to further increase a_D and thereby reduce the production time. The paper reports on the effect of the operating pressure (p) and a_D on the structure and electrical resistivity of Ti films which were deposited onto glass substrates held at the floating potential, by the magnetron sputter ion plating (MSIP) process, using an unbalanced d.c. magnetron. The deposition rate varied from 0.03 to 0.45 μm/min, depending on the magnitude of the discharge current I_d. The structure of Ti films, specifically the crystallographic orientation, grain size and microdeformation, were determined from XRD analyses. The results obtained indicate that the effect of decreasing the operating pressure during the sputtering of films onto substrates at the floating potential, is equivalent to the MSIP of films onto biased substrates at p of about 0.5 to 1 Pa, currently utilized in conventional sputtering. This is due to the increased role played by fast neutrals at low p. A further increase of a_D above 0.7 μm/min results in a dramatic change in the orientation of the crystallites from (002) to (010) when a particular threshold of target power density is exceeded. This result is of great practical importance.

1. *Introduction*

Understanding the effect of varying the deposition parameters on the development of the microstructure of deposited films is of extreme importance, as the microstructure plays a crucial role in the structural properties of the film. Varying the deposition rate, by changing the target current, results in changes in both the ion and coating particles energy and flux impinging onto the substrate. This in turn causes modified growth processes which result in changed microstructures. The purpose of this article is to report on the development of a new high rate sputtering magnetron under development in our laboratory, and on the initial analyses of films deposited in it.

2. *Experimental details*

The Ti films where deposited in a stainless steel chamber (diameter 150 mm and length 195 mm) using an unbalanced (UB) planar magnetron equipped with two electromagnets, supplied by the currents I_1 (internal) and I_2 (external) (see Fig. 1). The chamber was pumped down by an oil diffusion pump (2000 ls^{-1}), backed by a rotary pump (60 m^3h^{-1}). A pure Ti (99.5%) target (diameter 100 mm) was used to produce films onto glass substrates (36mm × 26mm × 1 mm), held at the floating potential U_{fl} at argon (99.99%) pressures from 0.06 Pa to 1 Pa and distances substrate to target d_{s-t} 50 and 80 mm. The thickness of the films, h, ranged between 0.8 and 4 μm. The deposition rate a_D was calculated by dividing h by the deposition time, t_d. Resistivities were measured by the four-point probe method at room temperature immediately after the films were removed from the deposition chamber.

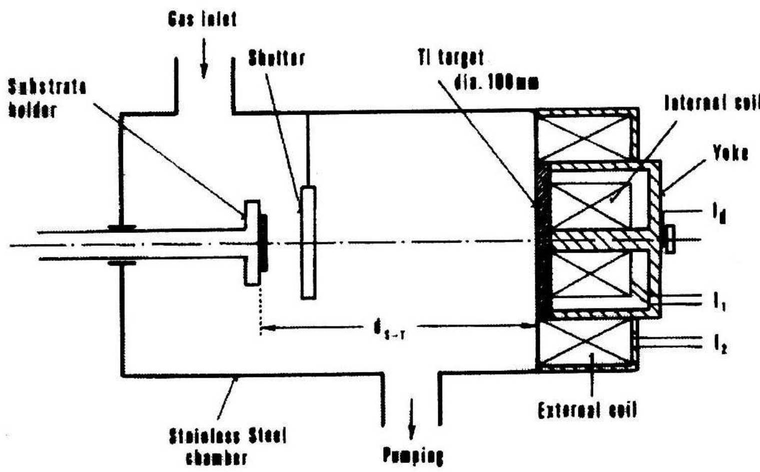

Fig. 1. Scheme of the deposition apparatus.

The thicknesses were measured by an Alphastep 100 profilometer, whilst the XRD analyses were performed on a Siemens D-500 diffractometer, using filtered CoKα_2 irradiation.

3. Results and discussion

3.1. Operational characteristics of an unbalanced magnetron

To achieve efficient magnetron operation (i.e. low pressure operation, lower power densities, etc.), it is necessary to ensure that the maximum plasma confinement possible, above the magnetron target (cathode), is attained. In our research magnetron sputtering device, this is achieved by varying the internal (I_1) and external (I_2) currents in the electromagnetic coils of the magnetron (Fig. 1). The variations in the currents leads to changes in the magnetic field configuration above the cathode and, hence, to changes in the plasma confinement. If this confinement (plasma density) is increased (i.e. the density of charged particles), then the deposition rate a_D is increased (due to increased ion/target collisions which increase the probability of an atom to be sputtered) and the minimum operating pressure of the magnetron is lowered [1,2]. The film properties and microstructure can also be affected by a variety of parameters, including deposition rate, target power density W_T, distance substrate to target d_{s-t}, substrate temperature T_s, substrate bias U_s and operating gas pressure [3-9]. Via different mechanisms, these parameters affect the energy available to the adatoms for migrating and coalescing on the substrate surface, and hence result in the formation of films with different microstructures and attributes. It is therefore of immense value to study the plasma confinement which can be characterized by the ignition, p_i, and the extinction, p_{ex}, pressures of the discharge, shown in Figs. 2a and 2b. In this experimental set-up and for constant target current and external electromagnet current, $I_d = I_2 = 0.5$ A, an internal electromagnet current $I_1 = 1$ A is sufficient for maximum plasma confinement as there are no further variations in either p_i or p_{ex} (Fig. 2a). This means that, for this apparatus, a stable discharge can be ignited and sustained at the lowest possible pressures ($< 10^{-1}$ Pa) which increases the target utilization (lower operating pressures result in larger target erosion areas) and hence the efficiency of the magnetron. There is no dependence of p_i on the target current, I_d, (Fig. 2b) which indicates that there are sufficient numberes charge carriers (i.e. good ionization due to good plasma confinement) present to sustain the discharge current up to the investigated current $I_d = 5$ A. Also from Fig. 2b, it can be seen that for increasing I_d up to $I_d = 3.5$ A, there is an increase in p_{ex} from 0.02 Pa to 0.04 Pa. This increase of pressure is necessary to provide a more dense plasma (i.e. to increase the probability of charged particles being created) which can support the discharge current. The behaviour of p_{ex} for $I_d > 3.5$ A is a result of the design of the power supply, where the load in the circuit, to the detriment of the target, is now having most of the available voltage drop across it. This means that to sustain I_d, there has to be an even greater increase of the pressure and consequently of the charge carrier density which is observed. Eventually, even without the effects of the external circuit design, we would expect p_{ex} to approach p_i for increasing I_d for the same target dimensions.

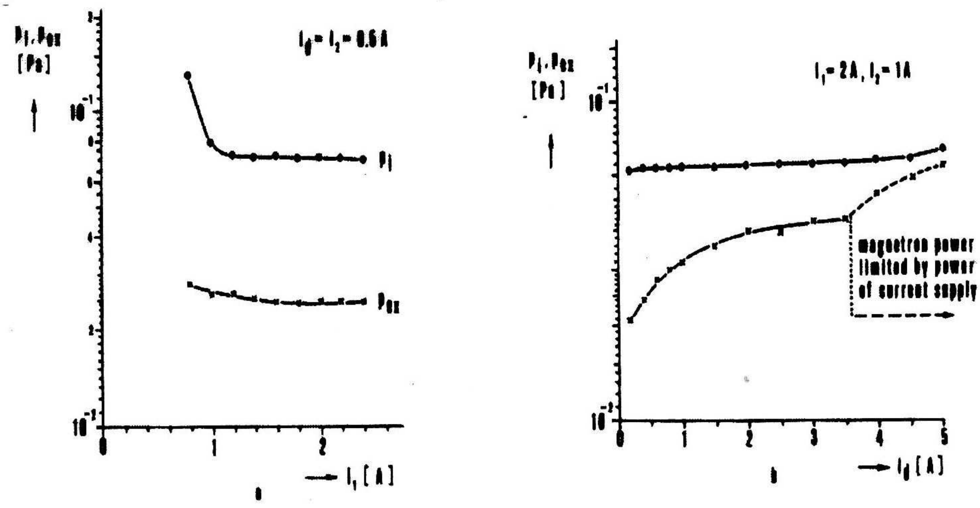

Fig. 2. Ignition pressures and extinction pressures as a function of a) internal electromagnetic coil current I_1 and b) target current I_d.

3.1.1. Deposition rate, a_D

When increasing the target power, P_T, up to $\approx$ 1500 W, there is an almost linear increase the deposition rate a_D, irrespective of the operating pressure, with a_D increasing as p decreases (see Fig. 3). This would be expected as at lower pressures and for low target power densities, the sputtered atoms have a longer mean free path and are, therefore, more likely to arrive at the substrate. Also, the racetrack (i.e. area of target sputtered) is greater at lower pressures for low target power densities and, hence, a_D is also larger. For $P_T > 1500$ W, the rate of increase begins to slow down. At the intermediate pressures, 0.15 and 0.4 Pa, the maximum deposition rate, $a_D \approx 0.34$ μm/min^{-1}, was achieved. The lowest and highest pressures have the lowest a_D. This behaviour of a_D for the lower pressure could be due to the increased loss of ions from the plasma, which would result in less sputtering of the target atoms. This loss of ions is caused by the expansion of the plasma due to rarefaction in front of the target caused by heat given from the target. At higher pressures, i.e. about 1 Pa, this heating would be less significant due to the fact that a lower voltage is needed to sustain the target current (i.e. lower target power is required) because of the increased density of charged particles. But this density itself will result in the sputtered atoms incurring more collisions, and hence a reduction in a_D will occur. The intermediate pressures result in a higher deposition rate, because they have a lower density of particles (i.e. fewer collisions occur than at 1 Pa) and a lower loss of charged particles due to plasma expansion (less heating than at 0.06 Pa). The method of calculating the deposition rate might also suggest that resputtering of the film could contribute to the observed decrease in a_D, but further research [10] shows that the decrease in a_D is not dependent on d_{s-t} (one would expect an increase in resputtering, which is synonymous with a decrease in a_D, if d_{s-t} is decreased, but that was not observed).

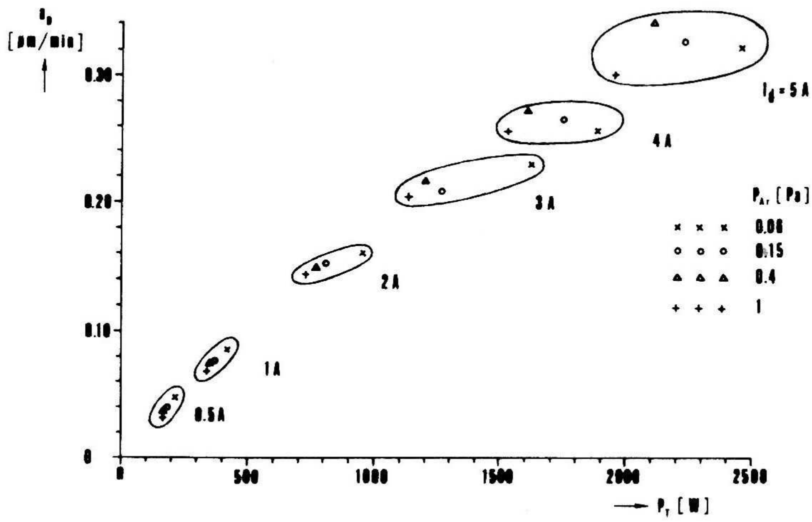

Fig. 3. Deposition rate a_D as a function of target power P_T for pressures 0.06, 0.15, 0.4 and 1 Pa. *Constant parameters:* $I_1 = 1.8$ A, $I_2 = 1$ A, $d_{s-t} = 50$ mm.

3.2. Characteristics and structure of the Ti *films*

3.2.1. Resistivity, ρ, of the Ti *films*

The decrease in resistivity for increasing I_d up to $I_d = 3$ A (Fig. 4) would be expected due to the increased energy being delivered to the film, which allows a defect free structure to be formed by encouraging surface and bulk diffusion processes [11-13]. For $I_d > 3$ A, ρ continues to decrease for the two highest pressures 0.4 Pa and 1 Pa as the energy supplied to the film increases. For the lowest pressures, 0.06 and 0.15 Pa, an increase in ρ is seen, caused by the domination of a second process, that of implantation damage due to fast neutral and increased energetic ion bombardment. This implantation damage will cause defects in the film to be formed, despite the increase in energy given to the film, and hence an increase in the resistivity will be observed (Fig. 4). As can be seen from Fig. 5a, discussed in section 3.2.2, there is no direct correlation between the resistivity and the grain size.

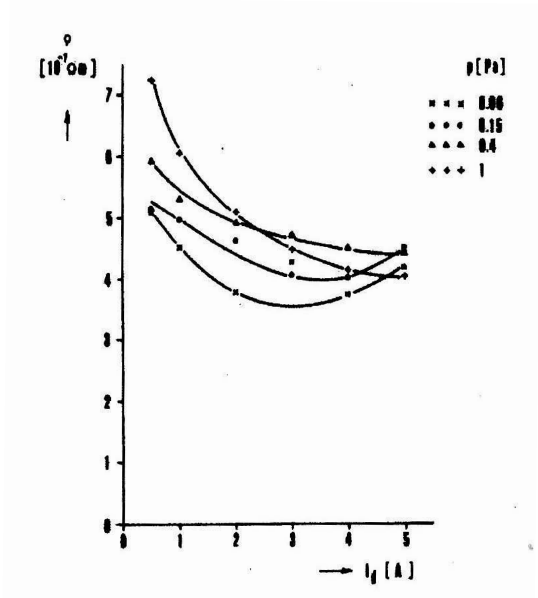

Fig. 4. Resistivity ρ as a function of target current I_d for pressures 0.06, 0.15, 0.4 *and* 1 Pa. *Constant parameters:* $I_1 = 1.8$ A, $I_2 = 1$ A, $d_{s-t} = 50$ mm, $h = 2$–4.8 μm.

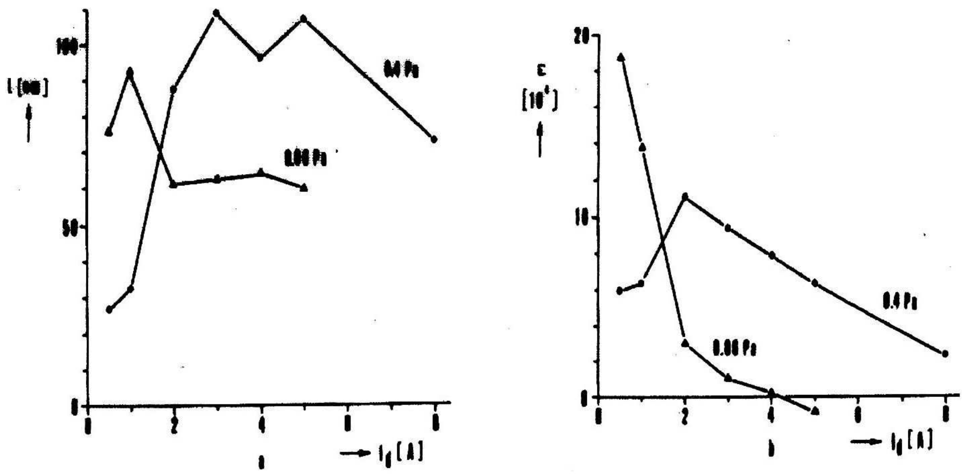

Fig. 5. The a) grain size L and b) microdeformation ϵ as a function of target current I_d for two pressures, 0.06 *and* 0.4 Pa. *Constant parameters:* $I_1 = 1.8$ A, $I_2 = 1$ A, $d_{s-t} = 50$ mm.

3.2.2. Grain size, L, of films

The grain size initially decreases when increasing I_d, and reaches a constant value of $L \approx 60$ nm for $I_d > 2$ A at $p = 0.06$ Pa (Fig. 5a). This behaviour of L is thought to be due to the increased energetic bombardment of the film by fast neutrals [7]. This could lead to formation of defects in the film which would result in a disruption in the grain growth by affecting the mobility of the grain boundaries and of adatoms [6]. At the higher pressure, 0.4 Pa, there is an increase in grain size for stronger $I_d < 3$ A, where $L = 110$ nm, followed by an almost constant value of L and then a decrease to $L = 70$ nm for $I_d = 8$ A (Fig. 5a). The increase observed at 0.4 Pa is due to the increase of the energy being delivered to the film which encourages the grain growth. The subsequent halt in the development of L is probably caused by a decrease in the ratio of coating particles to impinging low energy ions. This low energy ion bombardment neutralizes the grain growth which would be expected because of the increased energy being delivered to the film. For further increases of I_d, the bombardment will dominate over the heating process resulting in a decrease in L.

3.2.3. Texture of the Ti *films*

The degree of texture, T_i, of the films was characterized using the Harris texture index

$$T_i = \frac{I_i/R_i}{\dfrac{1}{n}\displaystyle\sum_{j=1}^{n}\frac{I_i}{R_i}}$$

where I_i is the observed intensity of the reflection $h_ik_il_i$, R_i is the corresponding intensity of a randomly oriented sample and n is the number of unique reflections (i.e., in counting a particular $h_ik_il_i$, the second orders should not be counted). When $T_i = 1$ for all reflections, than the sample is said to be randomly oriented. If $T_i > 1$ for some $(h_ik_il_i)$ planes, then it is said that the sample exhibits preferred orientation of the planes parallel to the film surface. In Fig. 6 the textures of the 010 and 002 planes are shown as other planes were barely present or are simply the second order reflections of 010 or 002 (Fig. 7). From Fig. 6, it can be seen that for increasing I_d up to $I_d = 18$ A, the 002 reflection gradually decreases whilst, simultaneously, the 010 reflection increases. For $I_d > 18$ A, an abrupt change in the texture from 002 to 010 occurs. The exact mechanism causing this change is not understood but is obviously linked to a change in the properties of the material from the target when a critical power density $W_{T_{Cr}} (> 120 \text{ Wcm}^{-2})$ is exceeded [14].

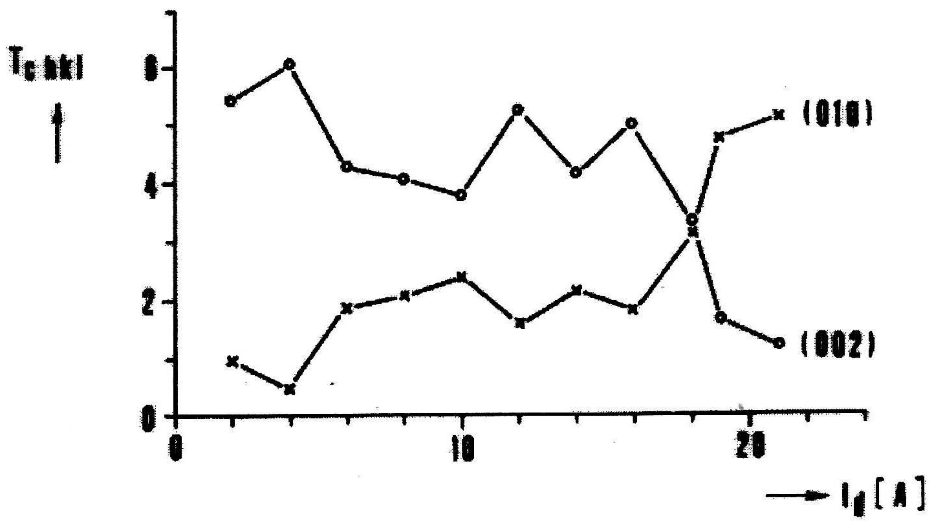

Fig. 6. Texture index T_i as a function of target current I_d. Constant parameters: $I_1 = 2$ A, $I_2 = 1$ A, $d_{s-t} = 80$ mm, $p = 0.4$ Pa.

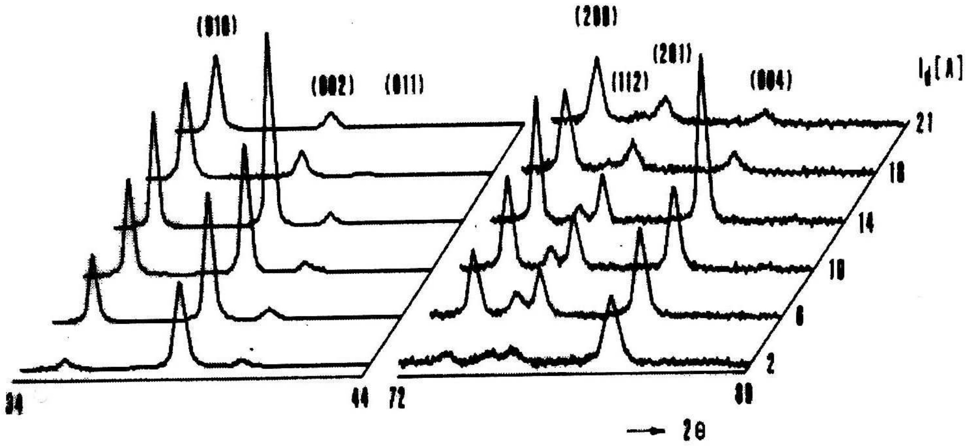

Fig. 7. Development of reflection $h_ik_il_i$ intensity for increasing target current I_d. Constant parameters: $I_1 = 1.8$ A, $I_2 = 1$ A, $d_{s-t} = 50$ mm, $p = 0.4$ Pa.

4. *Conclusion*

The experiments show that there is an almost linear increase in the deposition rate for increasing target power up to $P_T < 1500$ W, with a_D becoming progressively larger as the pressure is reduced. At $P_T > 1500$ W, the rate of increase of a_D slows down. The intermediate pressures, 0.15 and 0.4 Pa, result in a higher deposition rate ($a_D \approx 0.34$ μm/min) than those of 1 Pa and 0.06 Pa. This is thought to be due to the increased loss of charged particles as the discharge expands due to rarefaction, caused by heating in front of the target and a smaller number of particles available for the sputtering process at a lower pressure. But, when the sputtered atoms undergo more collisions, due to the greater density in the plasma, it is less probable that they will arrive at the substrate at the higher pressure. The result that intermediate pressures can lead to higher deposition rates than lower or higher pressures for similar target power densities could be of immense value in industry, where the search for reduced production times and economic efficiency is in progress. The simple reduction (or increase) in operating pressure could result in power savings and decreased production times.

It was also shown that when a certain critical target power density is exceeded ($W_{T_{Cr}} > 120$ Wcm^{-2}) then an abrupt change in the texture of the Ti films from predominantly 002 to almost exclusively 010 is observed. The changes in the texture are thought to be connected with changes in the process of target atom ejection. At a critical deposition rate, a_{cr}, pure sputtering changes into a combined process where both sputtering and partial evaporation at the target surface occur [14]. This implies that there is a physical constraint placed on the maximum (or minimum) deposition rate permitted (in these experiments $a_{cr} \approx 0.7$ μm min^{-1}) if films with a particular texture are required.

The resistivities of the Ti films decrease with increasing target current up to $I_d \approx 3$ A over the pressure range studied, with the greatest decrease in ρ occurring at the highest pressure of 1 Pa and the lowest resistivity ($\rho \approx 3.5 \times 10^{-7}$ Ωm) occurring at the lowest pressure of 0.06 Pa. Whilst the resistivities at the higher pressures (between 1 and 0.4 Pa) continue to decrease for $I_d > 3$ A, those at the lower pressures (between 0.15 and 0.06 Pa) begin to increase and continue rising up to $I_d = 5$ A, the maximum target current investigated. It is believed that the cause of this behaviour for $I_d > 3$ A is the intensity of bombardment of the growing film by fast neutrals, which are predominant at these lower pressures and high target currents, and result in defects being formed and smaller grain sizes in the film leading to the higher observed resistivity.

For $I_d < 1.5$ A ($W_T < 10$ Wcm^{-2}) both the grain size L and the microdeformation ϵ are greater in the films prepared at 0.06 Pa than those films produced at 0.4 Pa, whilst the converse is true for $I_d > 1.5$ A.

The results also indicate that the effect of decreasing the operating pressure during the sputtering of films onto substrates at floating potential and at higher target power densities, is equivalent to the MSIP of the films onto substrates at p of about 0.5 to 1 Pa, currently utilized in conventional sputtering.

Acknowledgement

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References

1) S. Kadlec and J. Musil, J. Vac. Sci. Technol., accepted for publication;

2) J. Musil, K. Rusňák, V. Ježek and J. Vlček, Vacuum **46(4)** (1995) 341;

3) M. J. Khandagle, S. A Gangal and R. N. Karekar, J. Appl. Phys. **74(10)** (1993) 6150;

4) J. Musil, A. J. Bell, M. Čepera and J. Zeman, Czech. J. of Phys. **45** (1995) 249;

5) K. L. Westra and D. J. Thomson, Thin Solid Films **257** (1995) 15;

6) H. Kheyrandish, J. S. Colligon and J. K. Kim, J. Vac. Sci. Technol. **A 12(5)** (1994) 2723;

7) J. Musil, A. J. Bell, M. Čepera and J. Zeman, for presentation at AMPT'95, August 8 - 12 1995, Dublin, Ireland;

8) H. Ueda, O. Kitakami, Y. Shimada, Y. Igari and I. Kusunoki, Jpn. J. Appl. Phys. **33** (1994) 2015;

9) S. Müller-Pfeiffer, H. J. Anklam and W. Haubenreisser, Phys. Stat. Sol. (b) **160** (1990) 491;

10) J. Musil, A Rajský and A. J. Bell, unpublished;

11) M. Adamik, P. B. Barna, I. Tomov and D. Biró, Phys. Stat. Sol. (a) **145** (1994) 257;

12) M. C. Weinberg, J. Mat. Sci. **29** (1994) 5337;

13) M. E. Day, M. Delfino, J. A. Fair and W. Tsai, Thin Solid Films **254** (1995) 285;

14) J. Musil, A. J. Bell, A. Rajský, M. Čepera and J. Zeman, published in the Proceedings of The Third International Symposium on Sputtering & Plasma Processes 1995 Tokyo, Japan, 179.

UTJECAJ BRZINE NANOŠENJA NA STRUKTURU I OTPORNOST RASPRAŠENIH TANKIH SLOJEVA Ti

Istraživan je učinak radnog tlaka i brzine naparavanja na strukturu i električnu otpornost tankih Ti slojeva, nanešenih na neuzemljene staklene podloge magnetronskim i ionskim rasprašivanjem. Upotrebljen je neuravnotežen DC magnetron. Brzina nanošenja bila je izmedu 0.03 i 0.45 μm/min. Struktura Ti slojeva, veličina zrna i mikrodeformacije odredeni su rendgenskom difrakcijom. Povećanje brzine nanošenja iznad 0.7 μm/min vodi na izrazitu promjenu orijentacije kristalita od (002) ka (010) zbog premašenja praga gustoće snage.