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Unbalanced magnetrons and new sputtering systems with enhanced plasma ionization

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This paper presents a critical review of the present status and trends in the sputtering of thin films. A special attention is devoted to the magnetron sputtering, especially to unbalanced magnetrons (UM) and to new sputtering devices utilizing magnetic and/or electric plasma confinement. The last category of devices opens new possibilities in a production of high quality films. Sputtering systems using UM operated in the double-site-sustained discharge and in a closed multipolar magnetic field are described. The attention is also devoted to the effects of ion bombardment on the properties of the deposited films. In case of hard TiN coatings it is shown under what conditions dense compact porousless films can be created and how deposition conditions can influence stresses in films. At the end, several new systems which combine advantages of sputtering with other processes like the arc evaporation, the electron cyclotron resonance (ECR) plasma, and the laser irradiation are also briefly described.

I. INTRODUCTION

Today, the sputtering is one of basic, widely spread physical vapor deposition (PVD) techniques of thin films. To understand better main advantages, drawbacks, and problems which still remain to be solved, of the sputtering technique, we first shortly summarize a development of this technique. The development of the sputtering technique can be roughly divided into three steps: (1) a conventional diode or triode sputtering; (2) a magnetron sputtering (MS); and (3) a magnetron sputter ion plating (MSIP).

A real breakthrough in sputtering was the invention of the magnetron sputtering source,¹ in the early 1970s. In magnetrons, the charged particles are contained by a closed magnetic field and high density plasma is produced in the vicinity of the cathode. This results in (1) a strong decrease of the plasma impedance and a decrease of the discharge voltage, typically to ~ 500 V and (2) a strong increase of the deposition rate a_D , up to $\sim 1 \mu\text{m}/\text{min}$, of films created on substrates placed at typical substrate-target distances, $d_{s,t} \approx 50$ mm. Also, due to an excellent plasma confinement, the magnetron sputtering can be carried out at lower pressures compared to diode sputtering systems, typically at 0.1–1 Pa. Thanks to these advantages, the possibility to be operated in both dc and rf modes and also very simple and reliable construction of the magnetron source, the magnetron sputtering was introduced very fast into industrial utilization. In the beginning, the magnetron sputtering of thin films utilized only the flux of sputtered atoms. Substrates were placed outside the plasma and it enabled it to deposit films with a minimum substrate heating. The utilization of this technique made it possible to coat successfully thermally sensitive substrates, for instance, plastic foils with films of different metals.

Recently, it was recognized that it is also possible in magnetron sputtering to control the microstructure, chemical, and phase composition of the growing film when the plasma

is not strongly confined in the vicinity of sputtered cathode but rather the plasma is allowed to flow out from the cathode region in the direction of the substrate and the surface of the growing film is subjected to a flux of ions extracted from the plasma. Such a process is, according to Mattox,² called the ion plating and in the case of the magnetron sputtering, it is denoted as the MSIP.

To meet the requirements of MSIP, several new sputtering systems with enhanced ionization have been developed. The most important systems are the unbalanced magnetron (UM),³ the double site sustained discharge (DSSD),⁴ the magnetron operating with a magnetic multipolar plasma confinement (MMPC),⁵ two target sputtering device,^{6,7} the microwave electron cyclotron resonance (ECR) plasma sputtering device,^{8,9} and dual purpose cathodic arc/magnetron sputtering system.¹⁰ These and other new combined systems will be reviewed in this paper. Besides, we will describe some new construction of magnetrons, for instance, the compressed magnetic field (CMF) magnetron,¹¹ the interpole target-hollow magnetron (IPT-HM)¹² which is suitable not only for a sputtering of thick targets of magnetic materials but also for very high rate metal sputtering of about some thousand Å/s. Special attention is devoted to the sputter ion plating of films in large substrate-to-target distances and to the influence of the energy delivered to the surface of growing film during its growth.

II. SPUTTERING SYSTEMS WITH ENHANCED IONIZATION

In principle, an enhanced plasma ionization in sputtering systems can be achieved in two ways. These are based on (1) additional gas ionization and (2) plasma confinement.

A. Additional gas ionization

The additional gas ionization is used in sputtering systems equipped by conventional magnetrons (CM). It can be performed, for instance, using a hot cathode electron emission,¹³ [see Fig. 1(a)], or by using a hollow cathode arc electron source¹⁴ [see Fig. 1(b)]. In both systems a considerable increase of the ion current density i_s on substrates, placed in standard distances $d_{S-T} \approx 50$ mm was achieved compared with sputtering systems equipped with a conventional magnetron only.

The sputtering system with the hollow cathode has been used for a high rate sputter deposition of SiO₂ planarization layers¹⁵ and for development of low pressure straight-line magnetron sources for the lift-off technique.^{14,16}

B. Plasma confinement

An appropriate shape of the magnetic field together with electric fields can confine the plasma of the magnetron discharge and increase the gas ionization at substrates, even without any auxiliary source of ionization. Several magnetic field configurations have been developed: (1) UM sputtering (Ref. 3); (2) magnetron sputtering using a MMPC (Ref. 5); and (3) two targets sputtering with electric mirrors.^{6,7}

The principle of the UM is shown schematically in Fig. 1(c). In this magnetron the CM magnetic field is modified in such a way that the magnetic field lines, emanating from the

edges of the magnetron target, are directed to the substrates. Therefore, a dense plasma is confined by the magnetic field between the magnetron target and substrates.

If the magnetic field reaching substrates is strong enough, the UM can be operated in a new discharge mode, called DSSD.^{4,17} This operation mode makes it possible to transport high ion currents to substrates placed even in large distances, up to several hundreds of millimeters, from the magnetron target. The ion current extracted by the substrates in the DSSD mode is large and it can even exceed the magnetron discharge current.⁴

A fully different solution is the sputtering in a multipolar magnetic field. A multipolar arrangement of permanent magnets at the chamber walls ensures the MMPC in the sputtering system [see Fig. 1(d)]. This system makes it possible to confine large volumes of dense plasma.⁵ The greatest advantages of this system compared to the UM are (i) low magnetic field in the substrate area, high magnetic field near to the chamber walls and (ii) wide range of operating pressures down to 2×10^{-2} Pa.⁵ Typical operating parameters of CM, UM, DSSD, and MMPC sputtering systems are shown in Table I.

The two target sputtering systems, schematically displayed in Figs. 1(e) and 1(f) use for a plasma confinement the concept of combination of electric mirrors at two negatively charged magnets with an external magnetic field; the field lines of which connect both targets. The targets are either plane disks⁶ [Fig. 1(e)] or a ring-shaped target⁷ [Fig. 1(f)]. The plasma is confined between the two targets and the substrates can be placed either out of the plasma region to reduce the substrate bombardment by energetic particles or close to the plasma region if a plasma-assisted film growth is needed.

1. Utilization of unbalanced magnetrons for ion-assisted growth of thin films

The sputtering systems equipped by UM have become very popular systems for enhanced ionization. Since the first description of the UM in 1986,³ there has been a steadily increasing interest in both laboratory and industrial utilization of UM (see, for example, some recent review papers in Refs. 17–20).

In the last several years, according to our knowledge, about six laboratories (1) CSIRO Division of Applied Physics, Sydney, Australia;^{3,18,20,21–27} (2) Institute of Physics, Prague, Czechoslovakia;^{4,5,17,28,29} (3) Loughborough University of Technology, United Kingdom;^{30–33} (4) BIRL Northwestern University, Illinois, USA;^{34,35} (5) D. G. Teer Coating Services Ltd., Hartlebury, United Kingdom;^{36–38} and (6) Hauzer Techno Coating, Venlo, Holland³⁹ have become intensively engaged in ion-assisted sputtering of thin films using UM. The first four laboratories are engaged mainly in basic research, and the last two companies in industrial applications of UM.

Window and Savvides studied the ion fluxes ν_i from UM (Refs. 21 and 22) and investigated the effect of low ion energy bombardment on the properties of deposited films and on a modification of the film microstructure, mainly the generation of stresses and strains, preferred orientation, and the

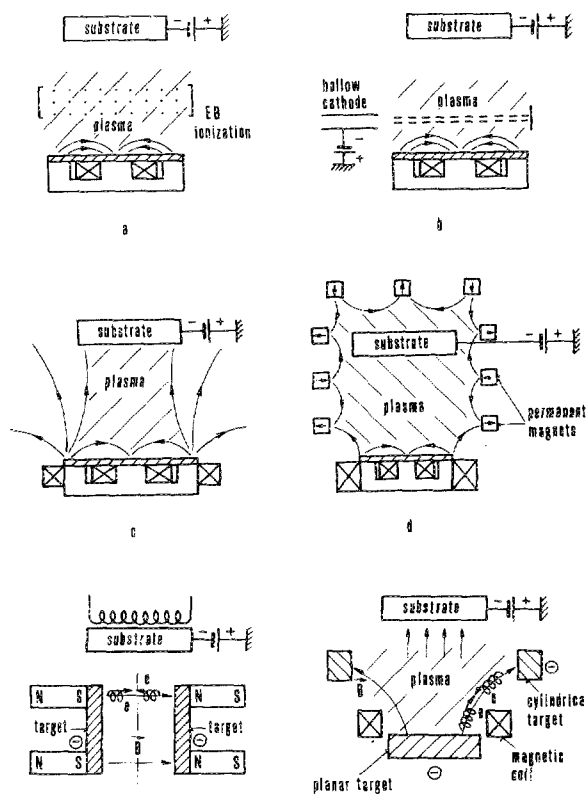


FIG. 1. Schematic illustration of sputtering systems with (1) additional gas ionization using (a) CM and the hot cathode electron emitter (Ref. 13) and (b) CM and the hollow cathode arc electron source (Ref. 14) and (2) improved plasma confinement (c) UM; (d) magnetron sputtering with MMPC (e) and (f) two targets sputtering system in external magnetic field.

TABLE I. Examples of typical operating parameters of magnetron sputtering systems with plasma confinement for moderate discharge powers.

Sputtering system	Magnetic induction B		Ion current density i_s		Distance d_{s-T}	Pressure p_T
	Substrates (mT)	Walls (mT)	Value (mA/cm ²)	Area of homogeneity	(mm)	(Pa)
CM	0.1	0.1	0.5	Large	50	0.1–1
UM	0.5	0.1	0.5–2	Small	50–100	0.1–1
DSSD	5	1	1–10	Small	50–500	0.05–10
MMPC	2	100	1–10	Large	50–500	0.02–1

effect of plastic flow.²⁶ The ion energy varied in the range 2–200 eV and the v_i/v_a atom arrival ratio in the range of 0.4–10. Their investigation was concentrated mainly on a -C film (Ref. 22), TiN films (Refs. 22 and 24) Ni/Cr alloy film (Ref. 22), B1 phase MoN superconducting film (Ref. 23), Mo and Zr films, (Ref. 25 and 26) W films,²⁶ and highly oriented YBa₂Cu₃O_{7-x} high T_c superconducting films.²⁷

Similar studies were carried out also in the other laboratories. Musil, Kadlec, and co-workers investigated mainly TiN films.^{5,17,28,29} They developed new sputtering devices equipped with UM capable of transporting high ion currents to substrates placed at large distances d_{s-T} and to coat large substrates of complex shapes.

Howson and co-workers^{30–33} studied the use of UM for a deposition of oxide layers as in In₂O₃ and TiO₂. The UM served as a source of plasma with a high electron temperature producing high negative floating potentials U_f of substrates immersed in the plasma of about –10 to –60 V and a relatively strong ion bombardment of dielectric films without any external bias.

2. Microstructure and stress in films prepared by ion-assisted magnetron sputtering

Many experiments carried out so far show the following important facts:

(1) The stress in the film sputtered at the lowest pressures is compressive and becomes tensile at higher pressures⁴⁰ due to thermalization of sputtered atoms and reflected energetic neutrals in collisions. The compressive stress in the film and its density increase with increasing ion bombardment^{20,41} and also depend strongly on the deposition rate a_D .²⁹ Ion bombardment at high negative biases $|U_s| \gtrsim 100$ V is usually accompanied by a resputtering effect.⁴

(2) Films in compression are compact, shiny, and exhibit dense, small crystallites microstructures. Films in tension are porous and usually are of a matt appearance.^{29,41}

(3) The stresses, strains, and microstructure of films deposited under ion bombardment can change from tensile to compressive, depending on the deposition conditions, mainly T_s , p_T , U_s , i_s , a_D , the deposited material, and the geometrical arrangement of the sputtering system.²⁹

(4) The compression-to-tensile stress transition in metal films moves to lower pressures with decreasing atomic mass of coating material⁴⁰ and increasing gas atomic mass.²⁵

(5) The compressive stress increases with increasing ion bombardment up to some maximum value when the material starts to flow and thus relieves some of the compressive stresses. Further increase in ion bombardment beyond this level resulted in a reduced level of final film stress.^{20,26}

(6) In ion-assisted sputtering the energetic sputtering atom and/or energetic neutral inert gas atoms reflected from the sputter target may be responsible for the creation of high compressive stress in the film produced at low pressures because of an atomic peening effect.^{25,26,42}

(7) The compressive stress in the ion bombarded film can be reduced by postannealing.⁴³

In some applications zero stress at room temperature is not desirable. For example, experiments indicate that hard coatings should be deposited in such a way to be under extremely high internal compressive stresses at room temperature.⁴⁴ In case the thermal expansion coefficient of the film is lower than that of the substrate, hard coatings should be deposited at a high temperature to achieve a reduction of the stress during the tool operation.

C. Magnetron sputter ion plating of hard coatings at large substrate-to-target distances

Until 1989 it was difficult to sputter good quality hard wear resistant TiN coatings on large tools and mechanical parts, mainly due to the fact that only low ion currents ($i_s \lesssim 0.5$ mA/cm²) could be extracted by a substrate placed at large d_{s-T} and that the ion cleaning of the substrate surface and the ion plating process were very ineffective.

The first sputtering systems with enhanced gas ionization were operated at standard magnetron distances, typically $d_{s-T} \approx 50$ –100 mm. These systems were used for the sputter deposition of small tools like drills, frames for glasses, bracelets, watch cases, cutlery, and other objects by wear resistant or decorative coatings. The enhanced ionization in these systems was achieved by a suitable geometrical arrangement, i.e., by (1) CM operated at very short distances $d_{s-T} \lesssim 40$ mm (Ref. 17) and (2) magnetron twins⁴⁵ and magnetron trinity^{36,37} using on overlapping of two, three, or more plasma clouds.

One CM operating in a short distance d_{s-T} is suitable to coat thin plane substrates, for instance, cemented carbide inserts. The CM twins are suitable to coat, without a rotation, small tools of diameter up to ~ 10 mm placed in the

region of two overlapping plasm clouds. The CM trinity can coat cutting tools of < 220 mm diam.³⁶ A considerable increase in the film quality is expected to be achieved when CM are replaced by UM.

The new generation of sputtering systems, i.e., UM operated in DSSD and in MMPC, which ensure the enhancement of gas ionization near substrates even at large distance $d_{S-T} \gtrsim 100$ mm are based on a magnetic plasma confinement combined with electric mirrors (target and substrate). The sputtering systems can deliver large ion currents I_s (several mA/cm²) to substrates and can be operated in a wide range of pressures. These new sputtering systems can produce good quality coatings on large substrates, even of complex shapes, and are fully competitive with PVD coating machines based on the low voltage electron beam evaporation⁴⁴ or the cathodic arc evaporation process.^{46,47}

1. Industrial unbalanced magnetron

In 1989 Teer reported a development of the UM denoted PLASMAG and the cylindrical coating machine ($\phi 360 \times 1200$ mm) with three rectangular UM (110×1000 mm) azimuthally separated by 120° .^{36,38}

Recently, the Hauzer Techno Coating Company in Holland developed a new industrial unbalanced magnetron denoted as ABS-C-190 for the operation also at very large distances $d_{S-T} \gg 100$ mm. Typical dependencies of i_s , a_D , and v_i/v_a of this UM as a function of the substrate-to-target distance d_{S-T} are given in Fig. 2. The UM ABS-C-190 was successfully tested in the reactive deposition of hard TiN coatings. Good quality TiN films were prepared even at very large distances d_{S-T} of ~ 800 mm.³⁹

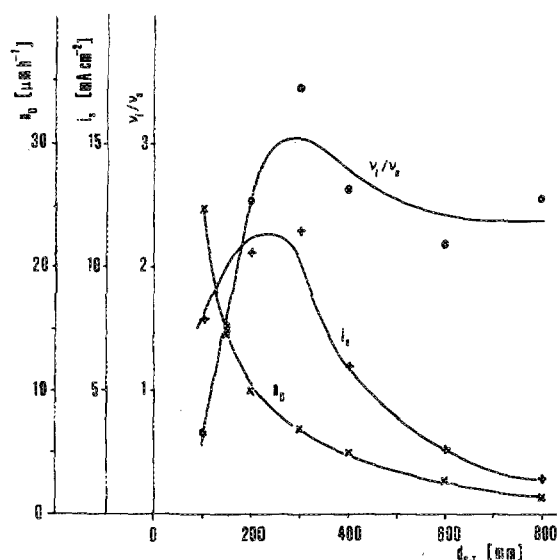


FIG. 2. Dependence of the deposition rate a_D , the substrate ion current density i_s , and the ratio v_i/v_a as a function of the substrate-to-target distance d_{S-T} in a system equipped with the ABS-C-190 magnetron source. Argon pressure $p_{Ar} = 0.2$ Pa and the bias voltage $U_s = -50$ V (Ref. 39).

III. NEW MAGNETRON SOURCES WITH HIGH RATE SPUTTERING

A significant effort is continuously devoted to the development of new magnetron sources with a high sputtering rate capable of increasing the deposition rate a_D of sputtered films. The only practical way to do this is to increase the plasma density in the close vicinity of the sputtered target. In principle, this can be done either by decreasing the distance d_{S-T} or by strongly increasing the target power density to values of ~ 100 W/cm² or higher.

A. CMF magnetron

The CMF magnetron developed ~ 10 years ago,¹¹ was successfully used for the preparation of highly oriented ZnO (*c*-axis orientation) films which exhibited excellent thickness homogeneity of 3% over 20 mm substrate at pressures of $p_T = p_{Ar} + p_{O_2} = 27$ Pa, distance of $d_{S-T} = 17$ mm, discharge current of $I_d = 0.5$ A, and discharge voltage of $U_d = 360$ V.^{11,48,49}

The main advantage of this magnetron is a strong increase of the parallel component of the magnetic field along the target surface. External soft magnetic pole pieces were extracted above the target surface. This principle was later successfully used to overcome the problem of sputtering of magnetic materials like CoCr and NiFe with thick targets (up to 14 mm).⁵⁰⁻⁵²

B. Magnetrons operated at very high target power densities

Standard CM and UM magnetrons work with target power densities S_T typically ranging from 10–20 W/cm². The deposition rate a_D of films in sputtering systems using these magnetrons, depending on the sputtered material and composition of created films achieves values up to 0.1–1 $\mu\text{m}/\text{min}$. Such a_D are, however, insufficient for some applications.

A further increase of a_D can be achieved when the sputtered target is loaded by very high $S_T \gtrsim 100$ W/cm². Such systems are based on a good combined magnetic and electric plasma confinement in the vicinity of the target. A good plasma confinement near the target can be achieved in both the cylindrical post magnetron⁵³ and the planar magnetron, called IPT-HM.⁵⁰ In these systems the magnetic field lines are nearly parallel to the target surface. To achieve plasma confinement along the target, i.e., to prevent electrons from leaving the target surface the edges of the target are equipped with disks (post magnetron) or rings [IPT-HM (see Fig. 3)] overlapping the target and electrically connected with the cathode. In addition to a very high a_D , both systems exhibit very large utilization of the target surface, considerably higher than 70%. The postmagnetron with $S_T \approx 90$ W/cm² was used already in 1984 for high rate deposition of TiN films.⁵³ Recently, it was reported that IPT-HM magnetron can be operated in a self-sputtering mode if the target power density $S_T > 80$ W/cm², i.e., in the absence of sputtering gas.¹²

The sputtering systems equipped with these magnetrons

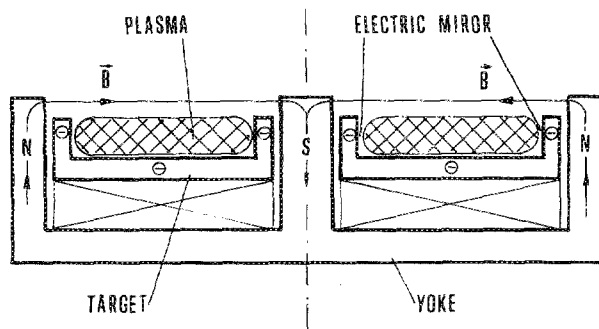


FIG. 3. Schematic principle of the IPT-HM (Ref. 50).

are, as to the film productivity, comparable to e -gun evaporating systems.

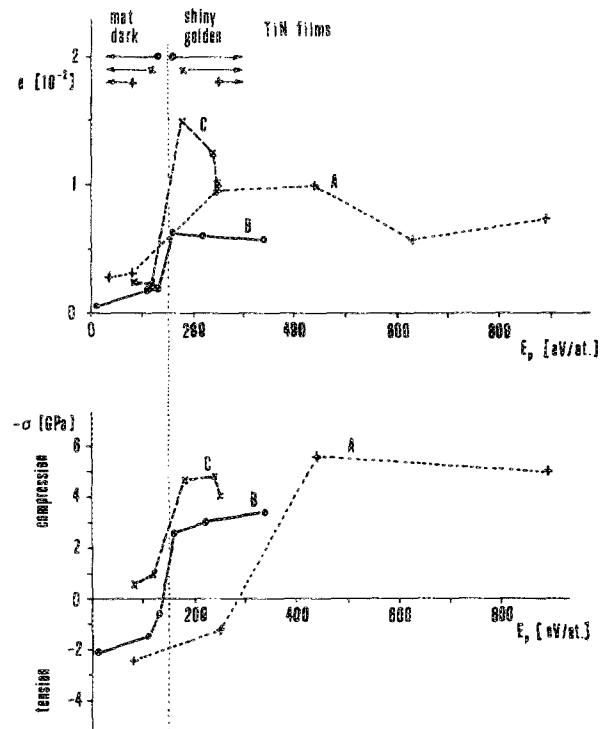
IV. ION ASSISTED FILM GROWTH

It is well known that the microstructure of deposited films and their physical properties depend substantially on the growth conditions. Basic microstructural features of deposited metallic films are described by structure zone models (SZM), developed for the evaporation as a function of T_s/T_m , (Ref. 54) CM sputtering as a function T_s/T_m , and p_T (Ref. 55) and bias CM sputtering as a function of T_s/T_m and U_s (Ref. 56). In these SZM, the film microstructure is classified into zones I, T, II, and III according to the parameters T_s/T_m , p_T , and U_s , where T_m is the melting point of the deposited film, p_T is the total pressure of sputtering gas, and U_s is the substrate bias.

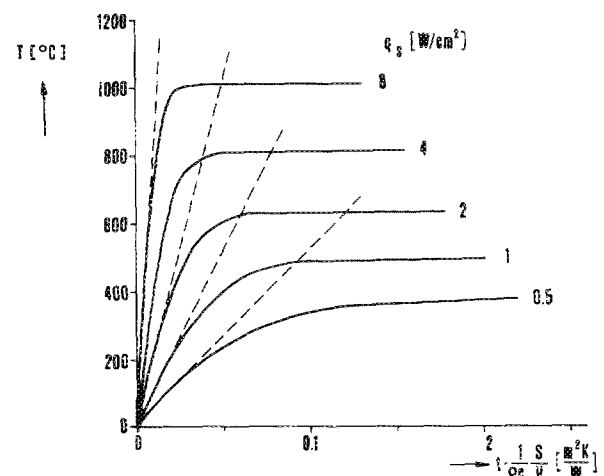
The newly developed sputtering systems with enhanced ionization have made it possible to vary the ion current density i_s independently of the deposition rate a_D and vice versa. The influence of both i_s and a_D on the film microstructure and properties can therefore be studied. In recent papers on reactively sputtered TiN films^{4,5,29,57} such effects have been studied. Sharp transition in the film microstructure from porous (zone I) to dense (zone T) occurred in any of the following parameter changes: (i) increase of U_s ; (ii) increase of i_s ; (iii) decrease of a_D ; and (iv) decrease of total pressure p_T , for each change the other parameters being kept constant.

These results support the hypothesis that, in the ion plating process, the film microstructure, mainly the zone I/zone T boundary, could be controlled by the combined parameter $E_p = eU_i v_i / v_a \propto U_s i_s / a_D$,^{17,29} where e is the electron charge, U_i is the energy of ions, v_i is the ion flux, and v_a is the sputtered atom flux at substrates. In fact, E_p is the average ion energy per one deposited atom.

The microstrain ϵ and macrostress σ of stoichiometric TiN films as a function of E_p are shown in Fig. 4 for three sets of films, A, B, and C. The independent parameter varied in deposition was i_s in set A, U_s in set B, and a_D in set C. For all three sets of films the transition from porous and mat dark (zone I) to compact and shiny golden (zone T) films occurs when the parameter E_p increases over ~ 150 eV/atom. The results indicate that in future structure zone models the parameter E_p could play an important role.

FIG. 4. The microstrain ϵ and the macrostress σ in TiN films as a function of the ion energy per condensing atom E_p . Independent variable parameter was i_s in set A, U_s in set B, and a_D in set C of TiN samples (Ref. 29).

The above-mentioned facts have serious implications on thin film deposition processes. Particularly, there is often a practical need to increase the productivity in industrial processes, i.e., the deposition rate of films. Moreover, if the film microstructure should remain the same, the parameter E_p should be kept constant. It means that the power density of ion bombardment on substrates, $q_s = i_s U_s$, has to be increased in direct proportion to the increase of a_D . This results in an increase of the substrate temperature T_s (see Fig. 5). The substrate temperature T_s is however, often limited to a particular interval of T_s determined by the substrate prop-

FIG. 5. The substrate temperature T_s as a function of the time parameter for different values of the substrate power density q_s (Ref. 19). Parameters σ , c , S , and V are the density, the specific heat, the surface, and the volume of substrate, respectively.

erties and/or by the film growth requirements. Therefore, the substrate overheating has to be often prevented.

V. COMBINED PROCESSES

Recently, several new systems, which combine advantages of sputtering with other processes like the arc evaporation, the ECR plasma, and the laser irradiation, have been developed.

A. Combination of magnetron sputtering and cathodic arc evaporation

It is well known that cathodic arc evaporation source produces a flux of highly ionized metal which is beneficial for the creation of good quality coatings. Unfortunately, it suffers from a generation of macroparticles which incorporate into growing film. On the contrary, this drawback is absent in the magnetron sputtering. Therefore, a new dual purpose cathodic arc/magnetron sputtering system has been developed^{10,39,58} so that one source can be operated in both sputtering and cathodic arc evaporation modes.

The process starts with cathodic arc evaporation which is used to bombard the substrate surface with metallic ions and creates an advantageous interface for deposition. Then the coating is deposited by magnetron sputter ion plating. In this way good quality TiN films with enhanced adhesion to substrates can be created.^{39,58,59}

B. ECR plasma sputtering system

In 1979 it was proposed to combine a microwave ECR plasma with sputtering or evaporation of materials to create good quality films at low substrate temperatures and low pressures of 10^{-1} – 10^{-3} Pa [Ref. 8 (see Fig. 6)]. This system can be operated in a mixture of inert and/or reactive gas and material vapors which is ionized in the ECR region. In this way created plasma can be accelerated in the direction of decreasing magnetic field to substrates.^{60,61} The energy of ions bombarding the growing film depends on T_e , grad B , and the shape of the magnetic field B in a space between the ECR region and substrates. For more details, see Ref. 61.

Due to (i) high degree of plasma ionization ($\geq 10\%$); (ii) high plasma density of 10^{10} – 10^{12} cm $^{-3}$; (iii) relatively low $T_e \leq 10$ eV; (iv) possibility to control the ion energy in range from of ~ 10 to several tens eV,⁶² this system permits us to synthesize new high-quality thin films at low temperatures and high deposition rates.

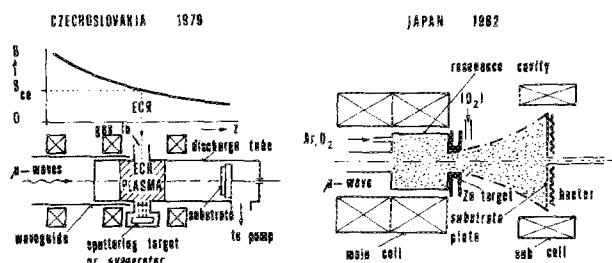


FIG. 6. Schematic of ECR plasma sputtering device for thin film processing utilizing principle of plasma acceleration: (a) proposed in Czechoslovakia (Ref. 8) and (b) developed in Japan (Refs. 9 and 60).

The ECR plasma sputtering system was already used for the deposition of different films as Al, Mo, Fe, Al_2O_3 , AlN, SiO_2 , ZnO, $YBa_2Cu_3O_{7-x}$.⁶³ Both insulator and conductor films can be prepared.

Recently, the ECR plasma sputtering system was improved by using two plasma enhanced sputtering targets with electric mirrors, according to Fig. 1(d).⁶³

C. Magnetron sputtering with concurrent laser irradiation

For a control of the microstructure, phase, and chemical composition of the growing film, not only ions and neutral particle bombardment can be used, but also photons, i.e., laser beam irradiation. Recently, for instance, a stimulated growth of ϵ -Ti $_2$ N phase in Ti-N films containing ~ 33 at. % N was experimentally demonstrated.⁶⁴

This combined system seems to be advantageous in cases when a generation of defects in the growing film should be reduced.

VI. CONCLUSION

At present the sputtering technique is widely used in both the basic research and the industrial applications. This technique and particularly ECR plasma sputtering systems already permits production of many high-quality materials at low temperatures like SiO_2 , Si_3N_4 , Al_2O_3 , Ta_2O_5 , ZnO, α -Si:H, Y-Ba-Cu-O, etc. In spite of this fact, further development of new sputtering systems goes on.

The development of new sputtering systems is stimulated mainly by (1) new knowledge achieved in the thin film physics; (2) the requirements of new applications; (3) the requirements of the industrial production of thin films, such as the low cost and the reliable operation of the sputtering device; and (4) the urgent need to deposit high-quality films with prescribed microstructure, phase, and chemical composition, i.e., with prescribed properties. There is no doubt that new more advanced sputtering systems will be developed in the near future.

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