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Recent aspects concerning DC reactive magnetron sputtering of thin films: a review

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Abstract

In this paper, attention is paid to DC reactive magnetron sputtering and its implications, such as the hysteresis effect and the instability in the reactive gas pressure, differential poisoning of magnetron cathode, as well as the methods which are used to control the process. These methods include: (a) increasing the pumping speed, which requires considerable additional costs; (b) increasing the target-to-substrate distance, which requires larger vacuum chambers (hence, higher costs) and also results in lower deposition rates; (c) obstructing reactive gas flow to the cathode with the resultant reduction of deposition rate and the need for complicated arrangements; (d) pulsed reactive gas flow, which requires an extensive amount of process optimisation and a continuous monitoring and adjustment of the process parameters; (e) plasma emission monitoring; and (f) voltage control, which are inexpensive and have proved to be powerful techniques for monitoring and controlling the reactive sputtering processes, in real time without disturbing the discharge, for the deposition of high-quality films, reproducibly. In addition, arcing and methods to avoid it are reviewed; this includes, arc initiations and their destructive effects (e.g. driving the process to become unstable, reducing the target lifetime and creating defects in the sputtered films), time required for arcs to occur, and finally, methods of avoiding arcs. The latter includes the use of unipolar or bipolar pulsing techniques at frequencies in the range 10–70 kHz. © 2000 Elsevier Science S.A. All rights reserved.

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1. Introduction

Reactive sputtering can be defined as the sputtering of elemental targets in the presence of chemically reactive gases that mass react with both the ejected target material and the target surface. It has become a very popular technique in today's search for new material properties, for the deposition of a very wide range of compound and alloy thin films including oxides, nitrides, carbides, fluorides or arsenides [1]. Besides the improved properties of non-reactively sputtered films, the popularity of DC reactive sputtering from elemental targets (i.e. a target consists of a single element) can be attributed to several factors, among them: (i) it is capable of producing thin compound films of controllable stoichiometry [2] and composition [3] at high deposition rates and on an industrial scale [4–6]; (ii) elemental targets are usually more easily purified, and hence, high-purity films can be produced [7]; (iii) the complexity and expense of RF systems can be avoided since metallic targets are generally electrically conductive, and hence, DC power can be applied; (iv) elemental targets are usually easy to machine and bond; (v) metallic targets are thermally conductive, which makes the cooling of these targets more efficient — thus, the range of the applied power can be extended (e.g. up to 50 W/cm^2 and higher) without the fear of being

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cracked; and (vi) films are deposited at temperatures less than 300°C [8].

Although reactive sputtering is conceptually simple, it is in fact a complex and non-linear process which involves many interdependent parameters [9]. The presence of the reactive gas at both the cathode surface and the substrate results in strong interactions of the reactive gas not only with the condensing material but also with the cathode surface, the so-called target poisoning. Reactions in the gas phase are ruled out for the same reasons that ions cannot be neutralised in the gas phase (there is no mechanism which can dissipate the heat of neutralisation to conserve both momentum and energy in a two-body system, therefore it can only happen at a surface). These cathode reactions are seen to increase suddenly at some rate of reactive gas flow. If flow control of reactive gas is used, such reactions are marked by a change in the impedance of the operating plasma, an abrupt increase in the system pressure (or more precisely, in the reactive gas pressure), a drastic drop in the deposition rate and a change in the film from metal-rich to gas-rich (i.e. a change in the stoichiometry).

2. The hysteresis effect and process instability

Fig. 1a shows schematically the well-known and important feature of reactive sputtering at a constant partial pressure of non-reactive gas, when flow control of reactive gas is employed; a hysteresis curve of the reactive gas partial pressure, P_r, as a function of the reactive gas flow rate, f_r, at a constant sputtering power (or as a function of sputtering power at a constant reactive gas flow rate). At low values of f_r (e.g. point A in Fig. 1a), almost all the available reactive gas is gettered at the condensation sites. As a result, no essential change in P_r is observed from the background level and the deposited film is metal-rich. This situation prevails until f_r reaches a critical value, f_{r1} (point B in Fig. 1a), where the flow rate of the reactive gas into the chamber becomes higher than the gettering rate of the sputtered metal. The reactive gas reacts, then, with the target surface to form a layer of the gas–metal compound. Sputtering rates from compound targets are less than that of pure metallic targets by a factor of 10–20, for mainly two reasons: (i) the sputtering yield of metal atoms from a compound on the target surface is less than that from a pure metallic target; and (ii) compounds have higher secondary electron emission coefficients than metals and hence most of the energy of the incident ions is utilised to breaking bonds with the resultant creation and acceleration of secondary electrons. Consequently, less metal atoms are sputtered and less reactive gas is consumed in the reaction, and a sudden and sharp rise in the reactive gas partial pressure to a new value, P_{r1}, occurs (point C in Fig. 1a). The deposited film is then gas-rich. If f_r is reduced following an increase in P_r to a high level, P_r will not decrease following the same trajectory as it increased. ΔP_r will stay constant until some value f_{r2} (point D in Fig. 1a) where it abruptly increases and the reactive gas pressure decreases to the background level. This is because the reactive gas pressure remains high until the compound layer on the surface of the target is removed and metal is exposed to be sputtered once more. As a result, the consumption of the reactive gas increases and the deposited film is metal-rich again. The dependence of both reactive gas consumption and deposition rate on reactive gas flow rate are seen in Fig. 1b,c, respectively.

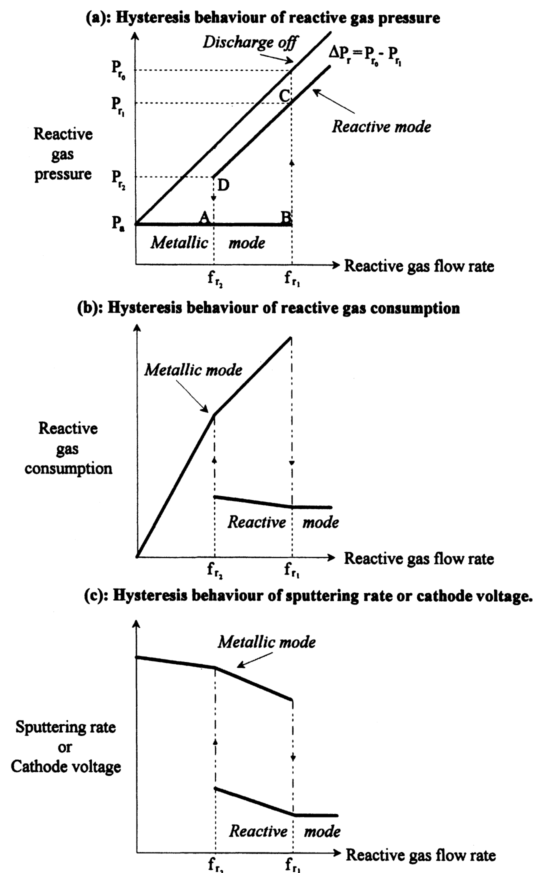

Fig. 1. The hysteresis behaviour of reactive magnetron sputtering.

The hysteresis effect is very undesirable and has to be eliminated, if flow control of reactive gas is used, because the process is unstable inside this region [10]. At one value of f_r, it is likely to deposit compound films of different stoichiometries and thus physical properties. This is due to the existence of two stable operating states corresponding to an individual value of f_r, when f_r is in the region of hysteresis. Fast and sophisticated process control systems are required in order to operate inside this region.

3. Differential poisoning of magnetron cathode

All the above discussion has been related to diode sputtering systems, which are characterised by a uniform plasma density across the cathode surface. This is not the case in magnetron cathodes, in which the plasma is highly localised in the racetrack region leading to an inhomogeneous discharge current density across the cathode. As a result, Schiller et al. [4,11] pointed out that the metal sputter rate, R_{sput}, and the oxidation rate, R_{ox}, are not constant over the target surface but show a noticeable site-dependence. Consequently, the degree of oxide coverage, Θ_{ox}, on the target surface must also be site-dependent leading to the possibility of the simultaneous existence of three states on the target surface; fully metal, fully oxidised or partially oxidised.

Fig. 2a represents a plot of $R_{sput}(x)/R_{sput}$ $(x=0)$ as a function of the distance, x, from the centre of the erosion zone. It appears that R_{sput} is highly site-dependent and shows a peak at $x=0$. Also shown in Fig. 2a, the fairly uniform distribution of $R_{ox}(x)/R_{ox}$ $(x=0)$ as a function of x. This means that at a moderate O_2 partial pressure, p_{O_2}, $R_{sput} > R_{ox}$ in the centre of the erosion zone and the difference between these two values decreases as x increases until a certain point where $R_{sput} < R_{ox}$. It should be mentioned that even in the erosion zone, oxides are continually formed and then destroyed.

If sputtering takes place in a very high p_{O_2}, and/or quite low discharge power, P, almost all the target surface will be covered by the reaction products (i.e. $\Theta_{ox} \approx 1$), as can be seen in Fig. 2b. Under these conditions, oxides are deposited onto the substrate with an

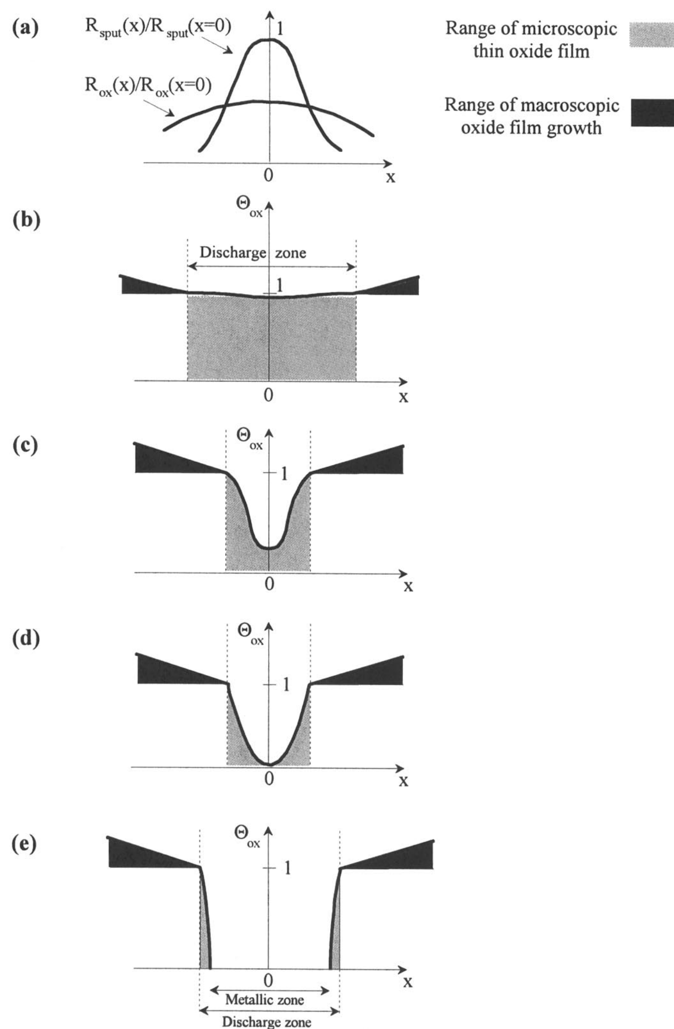

Fig. 2. Differential poisoning of magnetron cathode: (a) a plot of the sputtering rate, normalised to that at the centre of the erosion zone, as a function of the distance, x, from the centre of the erosion zone; (b) the coverage of the target surface with the reaction products at a very high oxygen partial pressure and/or low discharge power; (c–e) the gradual decrease in target coverage with decreasing oxygen partial pressure, and/or increasing discharge power. Reproduced from Schiller et al. [4,11].

extremely low rate due to the much lower sputtering yield of metal from an oxide target (Section 2).

With decreasing p_{O_2} and/or increasing P, Θ_{ox} decreases according to the distribution of the discharge current (or power) density within the discharge zone (Fig. 2c) until the target region corresponding to maximum discharge current density (i.e. where $x=0$) becomes entirely free from oxides (i.e. metallic with $\Theta_{ox} \approx 0$), as Fig. 2d shows. If p_{O_2} is further decreased and/or P is further increased, the width of the oxide-free zone on the target also increases, as shown in Fig. 2e.

Finally, adjacent to the metallic zone, on both sides, there are transition regions within which Θ_{ox} increases from zero to unity. Outside the transition regions a macroscopically thick oxide film then grows on the target surface.

Despite the differential poisoning at a given reactive gas pressure, the hysteresis effect, discussed earlier, was also observed in magnetron cathodes [8,9,12–14].

4. Methods of process stabilisation

For a deposition process to be stable, it is beneficial to operate the magnetron cathode with its surface either fully or partially metallic, while maintaining an adequately high partial pressure of the reactive gas at the substrate to form the desired compound film. This is not always possible because if the flow rate of the reactive gas is manually increased, the reaction between the reactive gas and the metallic target becomes inevitable. This in turn leads to the hysteresis behaviour, which is marked by an uncontrollable change-over (or instability) between a metallic and a poisoned target [13,15–18], exactly in the pressure range where a stoichiometric film is formed [19,15]. Methods of process control will be summarised below.

4.1. Varying the pumping speed

One solution to eliminate the hysteresis effect is simply to ‘overpump’ the deposition chamber [2,3,7,10,15,20–26]. This is achieved by providing a pumping system with a pumping speed that is sufficiently high so that the consumption of the reactive gas by the pumping system dominates that by the growing film. As a result, a gradual transition between the metallic (state I) and reactive (state II) modes takes place. Kadlec et al. [10] stated that during reactive sputtering, the reactive gas flowing into the deposition chamber, with a rate f_r, is either gettered by the sputtered material or removed by the pumping system. The equilibrium state is given by:

$$f_r = f_r^{sm} + P_r S_r \tag{1}$$

where f_r^{sm} is the flow rate of the reactive gas gettered by the sputtered material, P_r is the reactive gas partial pressure and S_r is the pumping speed of the pumping system for the reactive gas. Meanwhile, according to Spencer et al. [15], instability or hysteresis occurs when the target becomes poisoned at reactive gas pressures where a stoichiometric film is formed. Consequently, the consumption of reactive gas decreases due to the lower sputtering yield from compound targets. This leads to a surplus in the reactive gas (in the deposition chamber) causing its partial pressure to rise. The cycle that leads to instability or hysteresis is seen in Fig. 3. It can be concluded then that the deposition system becomes unstable if an increase in the reactive gas pressure results in a decrease in the reactive gas consumption (i.e. $\mathrm{d}f_r/\mathrm{d}P_r < 0$). For a stable system, the last inequality becomes: $\mathrm{d}f_r/\mathrm{d}P_r > 0$. Applying this condition to Eq. (1), one gets [24]

$$\frac{\mathrm{d}f_r^{sm}}{\mathrm{d}P_r} + S_r > 0 \tag{2}$$

If the critical pumping speed, S_c, is defined as [10]

$$S_c = -\frac{\mathrm{d}f_r^{sm}}{\mathrm{d}P_r} \tag{3}$$

the condition for the deposition system to become stable (i.e. to eliminate the hysteresis effect) is then

$$S_r > S_c \tag{4}$$

That is, the pumping speed of the pumping system should be greater than a critical value S_c. Spencer et al. [15] examined the above condition during the deposition of conducting films of indium oxide at a constant Ar partial pressure. They measured the O_2 consumption in the system when the magnetron was on (i.e. the total O_2 flow rate to both the growing film and the pumps), and when the magnetron was off (i.e. the O_2 flow rate to the pumps which was 240 l/s). From these measurements they obtained the consumption of O_2 by the film. Their results are reproduced in Fig. 4. The stability condition implied that $S_r > 150$ l/s. To examine this, they baffled the vacuum pump to give an O_2 pumping speed of 120 l/s. The results, which are reproduced in Fig. 5, indicated that the system became unstable with an uncontrollable rise in the O_2 partial pressure accompanied by a similar rise in the magnetron potential. The latter is a clear indication of a change in the state of the target (i.e. from metallic to oxidised). Furthermore, to prove that their results were

independent of their system or the material used, they carried out the same experiments in another vacuum system to deposit tin oxide. They reached the same conclusion, that is, instability occurs due to an insufficient pumping speed of the reactive gas by the pumping system. In the same paper, Spencer et al. [15] proposed a conceptual model of the reactive sputtering process. They first treated the effects of raising the reactive gas partial pressure at the target and the substrate independently, and then combined both effects to propose a model of the reactive gas consumption by the film. At the target, a partially poisoned surface results when the poisoning rate is higher than the cleaning rate. Since the sputtering yield of metal atoms from a poisoned surface is less than that from a pure metallic target, the metal flux from the target decreases with increasing reactive gas partial pressure until the entire target is poisoned, as can be seen in Fig. 6a. At the substrate, when the reactive gas partial pressure is low, the formation rate of the compound film is limited by the impact rate and utilisation of the reactive gas atoms and hence a metal-rich, or sub-stoichiometric, film is deposited. Since the impact rate of the reactive gas atoms increases with its partial pressure, the reactive gas-content of the compound film increases with the reactive gas partial pressure until a gas-rich, or over-stoichiometric (saturated), film is deposited (Fig. 6b). The combination of the processes, at the target and substrate, produces the curve of the reactive gas consumption by the film which is shown in Fig. 6c.

Okamoto and Serikawa [7] reactively sputtered Si in an Ar/N_2 atmosphere using RF magnetron technique at a constant Ar partial pressure. The pressure of the sputtering gas was controlled by throttling a baffle valve located above the diffusion pump. They varied the pumping speed, by a factor of 20, by changing the flow rate of Ar from 5 sccm to 100 sccm. At 5 sccm, a well-defined hysteresis curve between nitrogen partial pressure and flow rate was obtained, and the deposition rate decreased from 15 to 3 nm. At 100 sccm, the

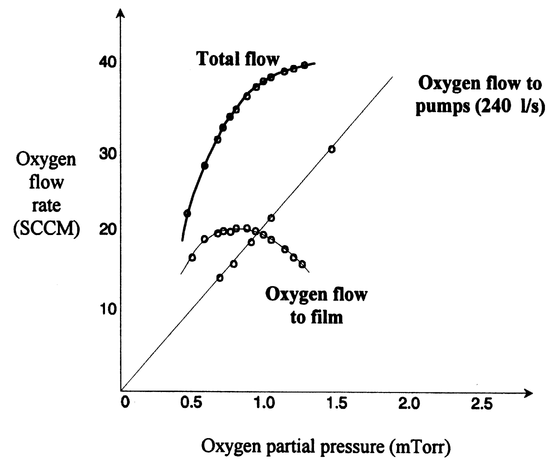

Fig. 4. The consumption of oxygen in a stable system depositing indium oxide. Reproduced from Spencer et al. [15].

hysteresis effect disappeared completely, although the deposition rate decreased to approximately 6 nm. However, the critical Ar flow rate was 62 sccm. Thus, the suppression of the hysteresis effect does not grant any significant benefits as far as the deposition rate is concerned. This conclusion was also reached by Berg et al. [20], who also found that the introduction of the reactive gas very close to the substrate (i.e. the creation of a gradient in the reactive gas partial pressure in the chamber [6,17]) decreased the poisoning of the target. They suggested this to be combined with a feedback control system, for the reactive gas partial pressure vs. the reactive gas flow rate, to be also able to operate within the instability where the most favourable conditions exist. These arrangements increase the deposition rate of compound films to a level virtually equal to that from metallic targets.

Finally, Danroc et al. [22] found, during the reactive magnetron sputtering of TiN, that the shape of the hysteresis curve changes as a function of both the

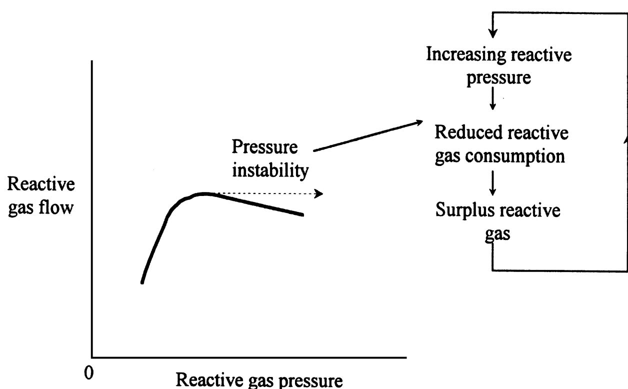

Fig. 3. The cycle that leads to instability in reactive sputtering. Reproduced from Spencer et al. [15].

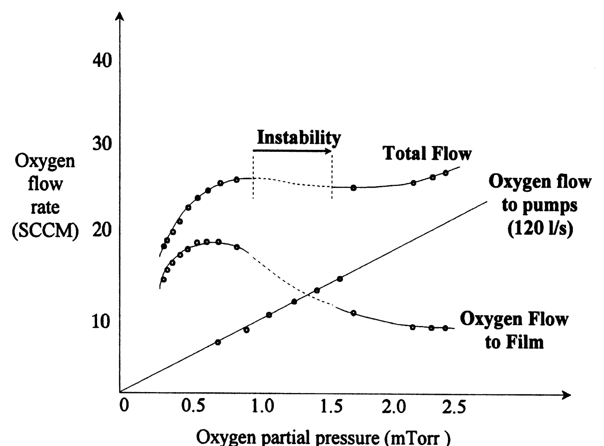

Fig. 5. The system of Fig. 4 made unstable by reducing the pumping speed. Reproduced from Spencer et al. [15].

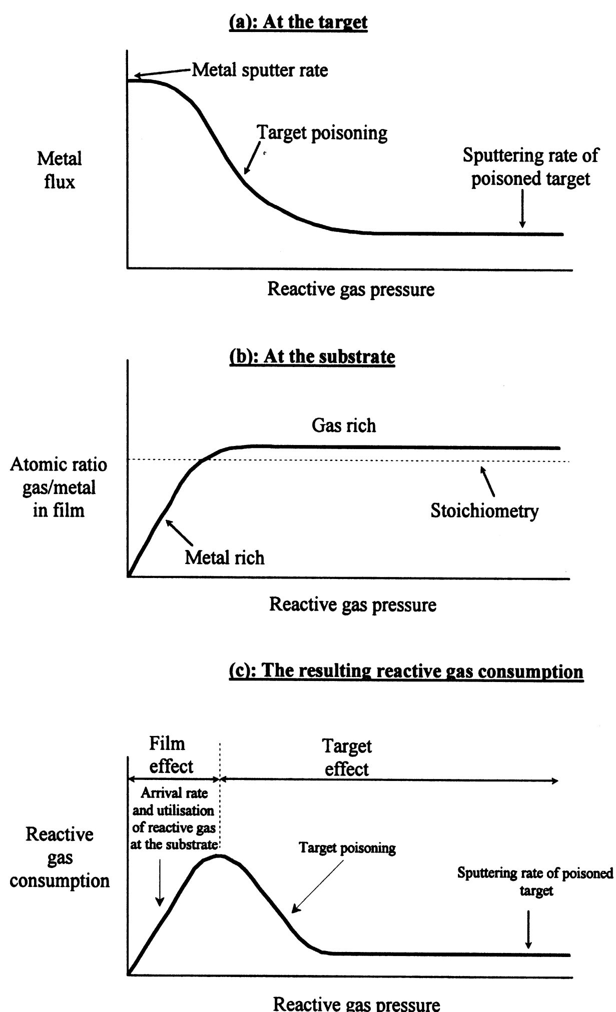

Fig. 6. The origins of the reactive gas consumption. Reproduced from Spencer et al. [15].

pumping speed and the position of the N_2 inlet. The width of the hysteresis curve decreased as the N_2 inlet was made closer to the turbomolecular pump entrance.

The problem with increasing the pumping speed, especially in industry where large vacuum chambers may be used, lies in adding considerable costs at the building stage of new machines, and can be difficult to adopt in existing ones.

4.2. Varying the target-to-substrate distance

Another method to reduce target poisoning is to make the target-to-substrate distance large. This effect was investigated experimentally by Schiller et al. [27] and published in a paper covering many important aspects of controlling the deposition process between the metallic and the reactive modes of the target. They found, during the DC reactive magnetron sputtering of Ti in an Ar/O_2 atmosphere, that when the target-to-substrate distance was small, over-stoichiometric or gas-rich films of $TiO_{2.4}$ were formed. This is because at such small distances, the flux density of sputtered Ti was high and hence a high O_2 partial pressure, P_{O_2}, was needed for the reaction to take place. The high P_{O_2} caused the target to switch to the reactive (or poisoned) mode, which is characterised by a low sputtering rate and gas-rich films. This indicates that optimum film properties can not be achieved from a poisoned target. When the target-to-substrate distance was made larger, the stoichiometric film of TiO_2 was formed at a lower P_{O_2}, and hence the probability of total poisoning of the target was reduced. The lower P_{O_2} is because the flux density of sputtered Ti was then lower due to the cosine distribution law.

In industry, varying the target-to-substrate distance is not practical because it requires considerably larger vacuum chambers, and hence, higher costs.

On the other hand, in the same paper Schiller et al. [27] investigated the variation of plasma emission intensity as a function of reactive gas flow rate. They found that the dependence exhibits a similar hysteresis curve to that of reactive gas partial pressure vs. reactive gas flow rate, as shown in Fig. 7, and concluded that plasma emission can be used to control the process within the instability. This important idea is not elaborated on until later papers by Schiller and his co-workers.

4.3. Obstructing reactive gas flow to the cathode

Maniv et al. [28] suggested a different solution to prevent magnetron target poisoning and hence hysteresis instability, while producing transparent conducting

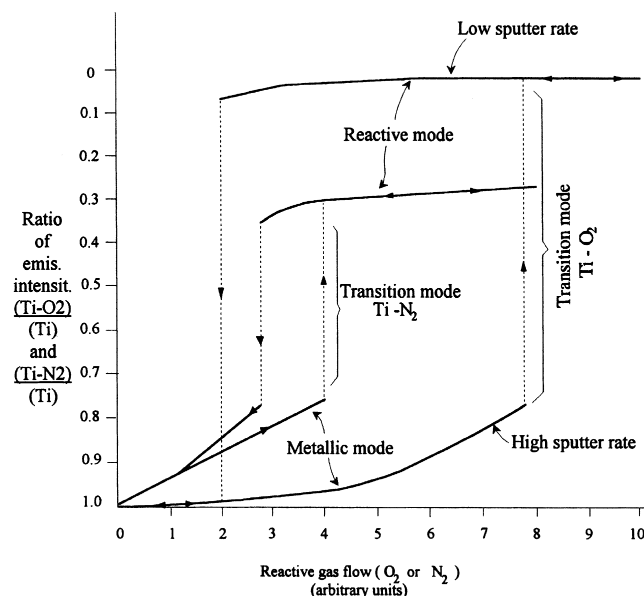

Fig. 7. Hysteresis in the plasma emission intensity in the wavelength range 460–470 nm with variation in reactive gas flow. Reproduced from Schiller et al. [27].

films of Cd_2SnO_4 by reactive magnetron sputtering of Cd_2Sn alloy target. They modified the magnetron system to reduce the oxidation rate at the target relative to the substrate by introducing a baffle between the substrate and the target, with O_2 inlet to the system on the substrate side, as can be seen in Fig. 8. The baffle also provided additional gettering surfaces for O_2 in the proximity of the target [15]. With this arrangement, it was possible to deposit stoichiometric films on RF-biased substrates before oxidation of the target was complete (i.e. before transition from state I to state II takes place). Maniv et al. [29] also used a baffle to obtain transparent conducting ZnO without RF coupling to the substrate. Many workers reported the successful use of this technique for the production of compound films. For example, Scherer and Wirz [30] for Al_2O_3, SiO_2, SnO_2 and Ta_2O_5, Este and Westwood [31,32] for Al_2O_3, AlN and TiN, Czternastek et al. [33] for ZnO doped with In (ZIO), Brett et al. [34] for ITO and ZnO and Brett and Parsons [35] for ZnO. However, the necessity for frequent cleaning of the baffle, the reduction of the metal flux to the substrate and the reduction of plasma-bombardment of the substrate, if the baffle is grounded, are the principal drawbacks of this technique. The last problem may be solved by either placing a positively-biased electrode in the substrate region to pull the plasma out of baffle, or by biasing the substrate.

4.4. Pulsed reactive gas flow

Aronson et al. [13] and later Sproul [36] proposed, during the DC reactive magnetron sputtering of Ti to deposit films of TiN, another method to maintain a stable operation in the metallic mode (i.e. at a constant N_2 partial pressure). They employed a pulsed reactive gas flow technique in which the reactive gas flow switches on and off periodically for short times. Two timers were used to independently control the pulse 'on' and 'off' times of a piezoelectric valve. The switching off is to allow the removal of any traces of compound formed onto the target surface before it accumulates and hence the target becomes irreversibly poisoned. Thus, the target switches between state I and state II, and the average target condition is then intermediate between the two states (i.e. the target continuously operating between the metallic and reactive modes around the knee of the hysteresis curve). Clearly, the pulsing time can not be too long otherwise the deposited film would be of alternate layers of compound and metal. Sproul [37] reported the successful deposition of TiN at a rate of approximately 50% of that of metallic Ti when N_2 was pulsed on and off at a rate of 3 s each. He also reported that the deposition rate of TiN, at the same cathode power, became virtually equal to that of metallic Ti when the pulsing rate was increased to 0.2 s. The pulse technique was also employed by Howson et al. [38] to deposit films of TiO_2. The main disadvantages of this technique are: (i) the need for an extensive amount of process optimisation prior to reproducibly depositing films with the required stoichiometry; (ii) the need for a continuous monitoring and adjustment of the process parameters. For example, it is not possible to deposit stoichiometric films at high rates unless the reactive gas flow is controlled manually to maintain a constant reactive gas partial pressure. Otherwise, either stoichiometric or over-stoichiometric films would be deposited at low rates accompanied by a rise in the reactive gas partial pressure, or sub-stoichiometric films would be deposited at high rates.

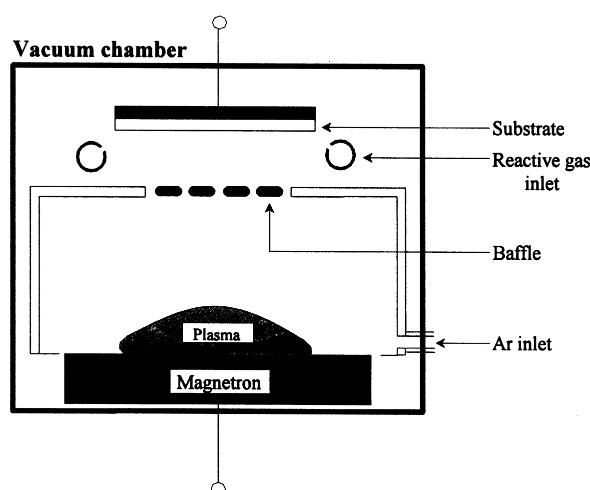

Fig. 8. Schematic of baffled magnetron sputtering system.

5. Methods of control to prevent instability

It is obvious that a stable operation is necessary to produce films of the desired stoichiometries at high deposition rates. This implies the need to operate at the knee of the hysteresis curve (i.e. in the transition mode at f_{r1}), which is usually very difficult to achieve because any major disturbance in the reactive gas partial pressure will shift the process to either extreme of the metallic or reactive mode. For example, any increase in the reactive gas partial pressure will drive the process to the reactive mode. In flow control, such a transition is irreversible since it will be impossible then to retrieve the desired set-point without tracing out the hysteresis curve, with the resultant deposition of inhomogeneous films. Consequently, a very fast feedback method has to be employed to automatically control the flow rate of the reactive gas into the system, or the discharge power, to maintain the desired set-point. This is accomplished by the continuous monitoring of any change in the operating point, using a process parame-

ter representative of the target state, and providing a feedback signal, in the control loop, to adjust either the flow rate of the reactive gas or the discharge power accordingly. Several parameters that continuously characterise the state of the process have been used as monitors. Cathode current, cathode voltage, reactive gas partial pressure, system pressure, deposition rate, film properties and light emitted from the plasma are the parameters which change rapidly near the critical flow f_{r1}, and have been used measure the process [17,37,39–49]. Unfortunately, not all these parameters allow the required degree of accuracy for process control. For example, reactive gas partial pressure is an equilibrium measurement, whereas in the case of controlling reactive sputtering, a transient process that keeps changing very rapidly when controlling within the instability region is confronted. Of these parameters, plasma emission monitoring (PEM) and voltage control techniques have proved to be very reliable in controlling reactive sputtering processes. Other methods of control, including successive plasma anodisation (SPA) and successive pulsed plasma anodisation (SPPA) will also be described.

5.1. Plasma emission monitoring

Although the use of light emitted from DC plasma discharges, as a diagnostic tool, is not new [50–53], the real re-birth of plasma emission monitoring was in the semiconductor industry for end-point detection in plasma etching [54]. After that, a US patent covering the utilisation of spectral emission lines from the plasma to control reactive sputtering in the region of instability was granted in 1979 [55], but the general use of this technique was very limited until 1987 when it was re-introduced by Schiller et al. [44] after they had alluded to it in 1984 [27].

Schiller et al. [56] have experimentally studied the relation between the emission intensity of the Ta line at 481 nm and the magnetron discharge parameters in a non-reactive atmosphere. They found that the emission intensity is virtually linearly proportional to magnetron current, magnetron power and deposition rate, at a given pressure. In addition, a strong pressure dependence was observed at given discharge parameters. In a reactive atmosphere [56], a pronounced drop in the emission intensity of the Ta line at 481 nm was observed as O_2 partial pressure increased due to the coverage of the target surface with the reaction products, which leads to a reduction in the metal sputtering rate. Furthermore, it was also observed that the emission intensity stays relatively high even if the O_2 content of the discharge gas is greater than 50%. Similar results to those of Ta were obtained using Al, Cu, Ti and In (90%)–Sn (10%) targets [56]. Blom et al. [16] also found, during the reactive deposition of ZrN films, that the nitrogen emission intensity increases linearly with the nitrogen flow rate.

It can be deduced from the above results, which have also been confirmed by many other workers [17,43,57–60], that the dependence of the emission intensity on the discharge parameters is evident. This made the emission intensity a valid control parameter to be utilised in controlling the reactive sputtering processes [44,55].

In PEM, the reactive gas inlet to the system is placed close to the target, the target-to-substrate distance is decreased and the creation of a plasma shield toward the chamber is encouraged. These arrangements improve the excitation of the reactants at the substrate, and hence, lower the flow rate of the reactive gas at which the desired degree of reaction occurs (i.e. f_{r1} and f_{r2} in the hysteresis curve are moved closer to each other). Thus, in contrast to the methods discussed earlier which tried to separate the event taking place at target and substrate, this method encourages such coupling in a way that the process would be unstable without it [39].

Spencer and Howson [45] developed a dynamic control for reactive magnetron sputtering to operate the magnetron in the instability region to produce films of better properties at higher deposition rates. They suggested that this can be achieved by breaking the feedback cycle (Fig. 3) through matching the reactive gas admission rate to the system and the consumption rate (by the film and the pumps) for the surplus of the reactive gas, which causes target poisoning, to be zero. A solution to the instability, in systems of limited pumping capacities [47], was obtained by providing a control of the partial pressure of the reactive gas with a rapid feedback. This was achieved by using an observation of the spectral line emission of the sputtered metallic atoms or the gas, in the plasma, as a signal representing the poisoning of the target or the partial pressure of the reactive gas in the chamber to control the admission rate of the reactive gas into the sputtering system [45,61]. Such a method of control allows the production of films of carefully controlled stoichiometries even at intermediate points that would be impossible to reach by ordinary pressure control apparatus.

Fig. 9 shows a PEM control loop. The light from the plasma is viewed through an optical fibre bundle. The bundle is directed into the centre of the magnetron to avoid deceptive indications of the target mode due to the simultaneous existence of three states across the magnetron cathode; fully metal, fully oxidised or partially oxidised (Section 3). The light is passed on to an appropriate optical filter (or a monochromator) which selects only the requested line emission. The selected wavelength is chosen so that the signal it passes (of the target material, the sputtering gas, or the reactive gas) is not influenced by the neighbouring lines of other

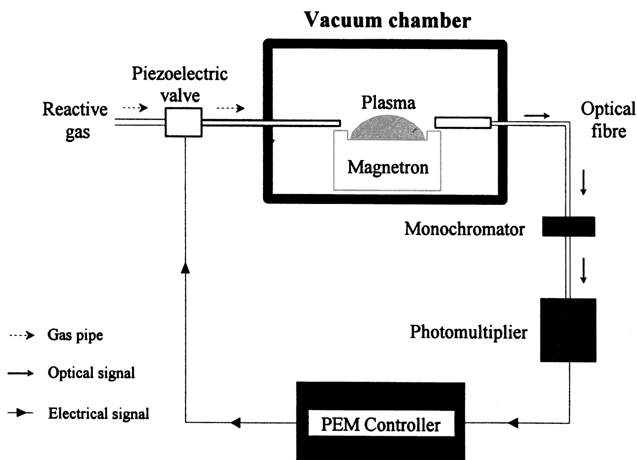

Fig. 9. Plasma emission monitoring (PEM) system used in controlling reactive magnetron sputtering processes.

elements in the plasma which may interfere with the desired signal. The obtained optical signal is converted into an electrical signal, proportional to the density of the corresponding element in the plasma, by a photomultiplier. The electrical signal is then applied to the PEM controller which is connected to a piezoelectric control valve of a very fast response. The controller compares the input signal to it with the desired set-point and gives the appropriate command to the piezoelectric gas flow control valve to admit/disallow a flow of the reactive gas to the system until the magnitudes of both the set-point and the input signal are equal.

PEM signals are sensitive to the sampling position, relative to target, and change with target erosion. However, these drawbacks can be overcome when the relative values of the signal, in a set of experiments, are considered. In fact, the absolute value of the PEM signal has no practical importance.

To conclude, the PEM is a powerful technique for monitoring and controlling the reactive sputtering processes for the production of high-quality films reproducibly. It samples from the region where the reaction is actually taking place, the information may be obtained in real time without disturbing the discharge and several peak emission intensities can be proportionally related to the sputtered metal flux and the partial pressure of the reactive gas [43].

5.2. Voltage control

An alternative parameter that continuously characterises the state of the reactive sputtering process and changes rapidly near the critical flow f_{r1} is the cathode voltage [33,49]. The cathode voltage is a measure of the degree of reaction at the cathode surface (i.e. target coverage) because it varies with the secondary electron emission coefficient from this surface, which is in turn sensitive to the surface conditions. For example, the secondary electron emission coefficients from oxide compounds are generally higher than their metal constituents (Section 2). Consequently, as these compounds are formed on the surface of the cathode, the cathode voltage will be changed. The cathode voltage may be measured and used to supply an electrical signal to the process controller to control the partial pressure of the reactive gas by adjusting the admission rate of the reactive gas to the system in a similar manner to that of PEM.

Voltage control is a less conventional method and may be employed in certain reactive sputtering conditions. These conditions are:

1. The difference between the cathode voltage of the metallic target material and that of the reaction product should be sufficiently large [55] and positive [21,62]. For example, the cathode voltage in the case of a pure Si target is ~ 700 V vs. ~ 200 V when SiO_2 is formed on the target surface, whereas the cathode voltage in the case of a metallic In (90%)–Sn (10%) target is ~ 385 V and the oxide is ~ 360 V. In the first case, the cathode voltage has been found to be a good indicator of the degree of reaction, whereas in the second case the range is too small and hence voltage control cannot be utilised.
2. The cathode voltage as a function of the reactive gas flow rate, or its partial pressure, should be single valued (e.g. In–Sn sputtered in Ar/O_2 is not) [55,62].

Voltage control has been successfully used by the author in the cases of Al, Zn, Cu and Pb when they were sputtered in an Ar/O_2 atmosphere.

5.3. Successive plasma anodisation (SPA)

An alternative to the simultaneous delivery of the constituent parts of a compound material (e.g. oxides) to the growing film, is to provide them separately, at different times, and react them together on the substrate [63]. Obviously, this can be achieved more easily on very thin layers of metal and the compound thin film can be built as an assembly of these.

In practice, this can be achieved by using two magnetrons, one to deposit a thin layer of metal and the other, which is strongly unbalanced, to oxidise the metallic layer in an Ar/O_2 plasma. The self-bias that appears on the isolated substrate, due to being immersed in the plasma that leaks from the unbalanced magnetron, helps in activating the reaction on the substrate by ion-bombardment and hence oxidising all of the metal. This process is repeated, to give the

total thickness of oxide that is required, by placing the substrate on a rotating table. A spinning rate of approximately one revolution per second is used.

5.4. Successive pulsed plasma anodisation (SPPA)

The SPA process, mentioned in Section 5.3, was developed to use just one unbalanced planar magnetron to both sputter the metal and oxidise it in Ar/O_2 plasma [63]. This was achieved by pulsing the admission of the reactive gas in response to a signal indicating the state of the cathode surface. In practice, an intermediate circuit was designed to obtain a minimum 0 V and a maximum 1 V setting corresponding to the signals proportional to the partial pressure of oxygen at the onset of fully poisoned and fully metallic target states, respectively. PEM or voltage control signals can be used. With the cathode operating in a fully poisoned mode, the output signal from the circuit to the reactive gas pressure controller was offset to 0 V. Then, with the reactive gas switched off and the cathode returned to the metal state, the output signal was set to 1 V. An electronic switching unit was designed to control the pulsing of the reactive gas into the chamber. Selection of two voltage threshold levels determined the open and closed states of a driver valve, which directed the reactive gas either to the process chamber or elsewhere. Admission of a reactive gas pulse to the chamber forces the signal below the selected 'lower threshold' (e.g. 0.2 V) and shuts off the reactive gas supply to the chamber, by diverting the flow, allowing the cathode to clean. The signal then rises above the 'upper threshold' (e.g. 0.8 V) where reactive gas is diverted back to the chamber and the target poisons once more. Repetition of this cycle was found to enable continuously variable and controllable target status.

With no reactive gas admitted, metal is sputtered. When oxygen is admitted, the target is poisoned and the just-deposited metallic layer is oxidised by the Ar/O_2 plasma. Again, the self-bias that appears on the isolated substrate, due to being immersed in the plasma that leaked from the unbalanced magnetron, helps in activating the reaction on the substrate by ion-bombardment and hence oxidises all of the metal. Howson [63] found that a switching time of approximately 1 s was appropriate for the deposition of a few atomic layers and their oxidation.

6. Arcing and methods of avoiding it

Highly insulating films (e.g. Al_2O_3, SiO_2 and TiO_2) have been traditionally deposited using RF diode or magnetron sputtering from compound targets [64–67]. However, besides the high cost of equipment, the power losses and the complexity in setting up an RF-powered sputtering system, the two principal disadvantages of RF diode sputtering are: (i) low deposition rates compared to DC sputtering; and (ii) films adhere to the composition of the target since the material of the target is in pressed powder form of well-defined composition.

The advantages of reactive sputtering from metallic targets (Section 1), have encouraged workers to think of alternative methods for the production of such films. The serious problem encountered during the reactive deposition of highly insulating films from metallic targets, using DC magnetron sputtering technique, is the build-up of an electrically insulating layer on the cathode surface with the resultant charge accumulation and arcing.

In the following sections, the causes of arc initiation will be summarised. The destructive effects of arcing on the deposition process, the coating properties and the target lifetime will be also discussed. Finally, methods of avoiding arcs will be described.

6.1. Arc initiation and their destructive effects

When a highly insulating compound is deposited, using DC reactive magnetron sputtering, both the metallic and the insulating states exist simultaneously on the cathode surface due to the differential poisoning of magnetron cathodes (Section 3). Since sputtering is a momentum transfer process, ion-bombardment of the cathode causes sputtering of both the insulating layers (Ins) and the metallic parts of the cathode (Met). However, unlike those that strike the metallic parts of the cathode, ions striking the surface of the insulating layers (Surf) cannot be neutralised by electrons from the cathode. Besides sputtering insulating layers, they only attract an equal number of electrons on the metallic surface of the cathode underneath the insulating layers, as can be seen in Fig. 10. From an electrical standpoint, the combination 'Surf-Ins-Met' represents a capacitor which keeps charging up as ions impinge on the surface of the insulating layer causing the voltage, V_{ins}, across the insulating layer to increase. Since the output voltage of the power supply is predetermined, the voltage across the cathode dark space should be reduced by V_{ins}, provided that the dielectric strength of the insulating layer is sufficiently high. This continues until the voltage across the cathode dark space becomes zero (i.e. the surface potential of the insulating layer is equal to the plasma potential). At this stage, no ions can acquire sufficient energy to further sputter the layer. However, if the dielectric strength of the insulating layer is not sufficiently high, the layer breaks down when the electric field across it reaches its dielectric strength. Consequently, an avalanche effect (i.e. a stream of electrons) occurs which results in a sharp

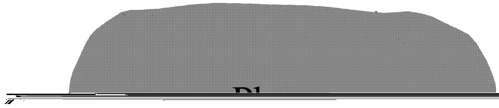

Fig. 10. Positive charge accumulation on the surface of an insulating layer.

increase in the current and hence temperature. The latter may produce local evaporation of target material [68].

On the other hand, any low impedance state (i.e. a high current and low voltage) observed at the output of the power supply caused by the process can be called an arc [69]. The most important sources of arcs at low gas pressures are field emission of electrons and macro particles [70]. Thermionic electron emission from the cathode as a result of heating it (e.g. cooling failure), outgassing from the cathode or the existence of a sufficiently high voltage gradient at the cathode [71] are among the probable causes of field emission of electrons. The other source of arcing is attributed to the accelerated macro particles which are detached from the cathode. If their energies are sufficiently high upon striking the anode, they cause local evaporation of its material. Due to the confinement of charged particles in magnetrons, these macro particles may end their lives at the cathode surface itself causing evaporation to its material.

It can be concluded from the above discussion, that a unipolar arc may be initiated by the breakdown of an insulating layer which builds up outside the racetrack of a magnetron cathode [72,73]. Schiller et al. [74] found that only 0.1% of these unipolar arcs may result in a bipolar arc. The undesired effects of arcing in reactive sputtering may be summarised as follows: (i) driving the process to become unstable [30,75–77] — this can be attributed to the disability of the system to recover its original state, after switching off the power supply temporarily to suppress an arc, due to the hysteresis effect [75]; (ii) reducing the target lifetime [69,73,74] — the target surface may be damaged due to the concentration of the entire discharge current in a very small spot of the cathode (with a diameter ranging from 1 μm to approximately 50 μm [74]) causing local melting of the target; and (iii) creating defects in the coatings [69,74–76,78–81] — this is due to the incorporation of droplets of molten target material, which are ejected from the target surface when an arc occurs, in the growing film. For example, arcing causes absorption losses in SiO_2 films prepared by reactive DC magnetron sputtering, because the emitted material usually contains a large number of metallic atoms which are not all completely oxidised [80].

6.2. Time required for arcs to occur

As has been stated in Section 6.1, the combination 'Surf-Ins-Met' represents a capacitor which continues to charge up as ions impinge on the surface of the insulating layer. The required time, t_B, to charge such a capacitor for the breakdown to occur, is given by

$$t_B = \varepsilon_r \varepsilon_0 \frac{E_B}{J_i} \tag{5}$$

where E_B, ε_r, ε_0 and J_i are the dielectric strength of the insulating layer, the dielectric constant of this layer, the permittivity of free space and the bombarding ion current density, respectively. Thus, breakdown can be prevented if the insulating layer can be discharged periodically with a period less than t_B, or a frequency greater than $1/t_B$. Eq. (5) indicates that t_B is dependent on the material involved and the deposition system [1]. It also indicates that t_B is independent of variations in the thickness of the insulating layer across the magnetron (Section 3). This implies that all regions of the insulating layer break down instantly. In magnetrons, however, the current density across the cathode is not uniform. It is higher near the thinner regions of the insulating layer (Section 3). Thus, according to Eq. (5), thinner regions break down first [69].

To get an idea about the approximate value of t_B, and hence the frequency, which are required to prevent arcing, the reactive magnetron sputtering of Si to produce films of SiO_2 will be taken as an example. In SiO_2, ε_r and $E_B = 3 \times 10^7$ V/m. If $J_i = 10$ A/m^2 in the border of the erosion zone [74], the discharging period, according to Eq. (5), should be $t_B < 100$ μs, which corresponds to a discharging frequency greater than 10 kHz.

On the other hand, Este and Westwood [82] studied the dependence of the sputtering rate on the frequency of the power supply, at a given power, during magnetron sputtering from Al targets. They measured the deposition rates at 60 kHz, 80 kHz, 500 kHz and 13.56 MHz and found that these rates were: 100%, 85%, 70% and 55% of that of the DC one, respectively. Consequently, the upper limit of frequency should not exceed 60–80 kHz. They also found that the deposition rate of AlN films at 100 kHz is 80% higher than that at 13.56 MHz.

As a conclusion, to secure an efficient avoidance of arc formation without affecting the deposition rate, the discharging frequencies of the insulating layers should be between approximately 10 and 70 kHz.

6.3. Methods of avoiding arcs

Tremendous efforts have been exerted by power supplies manufacturers to shorten both the detection time of an arc, and the switching off time of the power supply [81,83,84]. Although it helps to minimise the undesired effects of arcs, the rapid switching off of a power supply, upon detecting an arc, for a predetermined period of time followed by an automatic reignition of the discharge have been found to be insufficient to maintain the stability and the high deposition rate of the reactive sputtering processes of highly insulating materials [72–74,83]. As a result, attention has been focused in recent years on finding practical methods for preventing the formation of arcs instead of suppressing the already existing ones.

Arcing can be eliminated by not allowing such a degree of positive charge to accumulate, and hence, keeping the electric field across the insulating layer lower than its dielectric strength. In other words, arcing can be prevented by periodically discharging the charge build-up on the surface of the insulating layer before breakdown occurs. This can be achieved by the employment of the so-called 'pulsing technique', in which a low frequency power is utilised.

In the following sections, the two ways of applying the pulsing technique, namely: the unipolar and bipolar pulsing, will be discussed.

6.3.1. Unipolar pulsed magnetron sputtering

The basic arrangement for unipolar pulsed magnetron sputtering is shown in Fig. 11. The charge build-up on the surface of the insulating layer is restricted so that the electric field across the layer is kept below its dielectric strength. During the pulse-on time, sputtering of target material occurs and is accompanied by charge accumulation on the surface of the insulating layer. During the pulse-off time, the accumulated charge is discharged through the plasma (i.e. the charging potential is lowered via the plasma) [85]. For example, Frach et al. [76] utilised medium frequency pulsed power input into the discharge to reactively deposit films of Al_2O_3. A DC power supply was connected to the magnetron via an electronic switching device and a 'special network', as it was called, to periodically provide a pulse-on time for approximately 10–20 μs and a pulse-off time for approximately 10 μs. With this arrangement, a good process stability, a lower arcing frequency and a good film quality were attained.

Another example of effectiveness of this technique is the work carried out by Pond et al. [80]. They reactively

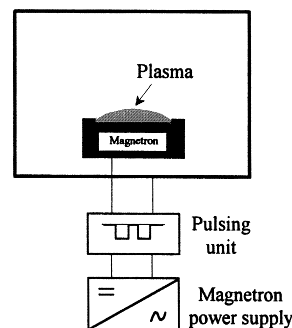

Fig. 11. The basic arrangement for unipolar pulsed magnetron sputtering. Reproduced from Szczyrbowski and Braatz [73].

deposited films of Ta_2O_5, Nb_2O_5 and SiO_2 as well as multilayer films of Ta_2O_5/SiO_2 (a 23-layer) to be used as an optical bandpass filter. They used the dual magnetron system which is shown simplified in Fig. 12. Since the reactive sputtering of Si in an Ar/O_2 atmosphere is accompanied by a severe arcing, the Si target was sputtered using a pulsed voltage formed by a modulator circuit and waveform generator, whereas Ta and Nb were reactively sputtered from the other target using normal DC voltage. The DC power supply was connected to a modulator circuit containing several switching transistors which switch the voltage from the power supply on and off according to the output of a waveform generator. The output voltage from the modulator to the Si target was modulated at 18 kHz with its most negative value equal to the output of the DC power supply, and its most positive value equal to zero. Again, good results, as far as process stability, arcing frequency and film quality are concerned, were achieved.

6.3.2. Bipolar pulsed magnetron sputtering

Instead of switching off the magnetron for a certain period of time to discharge the charged surface of the insulating layer via the plasma, as was the case in the unipolar pulsed magnetron sputtering, the surface of the insulating layer, in the bipolar mode, is discharged as a result of electron-bombardment when the polarity of the magnetron is reversed. Bipolar pulsed magnetron sputtering can be carried out using one magnetron or two magnetrons.

6.3.2.1. Bipolar pulsed magnetron sputtering using one magnetron. Historically, Cormia et al. [72] were the pioneers in using the low frequency AC technique in reactive magnetron sputtering. At such frequencies, the system behaves as if there were two cathodes with a dark space in front of each electrode. They deposited films of TiO_2. Using a conventional AC power supply, a 10-kHz AC potential was applied between a Ti target and a counter electrode which was, in their method, a conventional anode (a cylindrical bar along one edge of the target). During one half-cycle, the potential of the

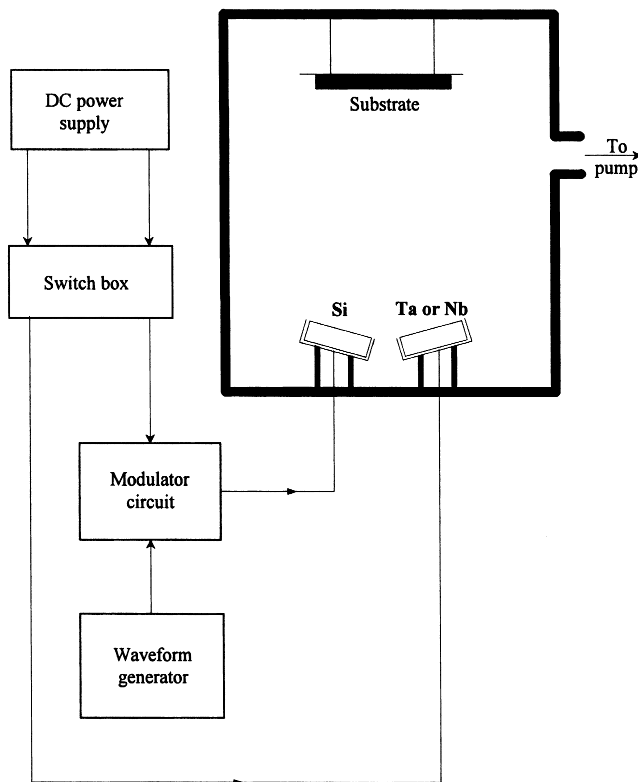

Fig. 12. Schematic of the modulated DC sputtering system. Reproduced from Thornton [78] and Williams et al. [79].

Ti target is more negative than the plasma potential and sputtering of its material occurs. In addition, the surface of the insulating layer is charged positively. In the other half-cycle, the surface of the insulating layer is discharged, due to electron-bombardment, as the polarity is reversed. The main disadvantage of Cormia's method can be explained as follows: to avoid arcing, ion and electron currents to the target must be the same (i.e. the average target current should be zero). As a result, a plasma has to be ignited even when the target is charged positively. Such a plasma is then sustained in the diode mode since the counter electrode, which now represents the cathode of the system, is a non-magnetron electrode. This requires, therefore, a very large positive voltage on the target on each half-cycle accompanied by sputtering of the counter electrode. In addition to the probable contamination of the deposited films, such an arrangement causes the electrodes to operate asymmetrically which results in an insufficient discharging of the surface of the insulating layer (e.g. SiO_2) [73] and hence arcing, although some improvement of process stability can be achieved.

Graham and Sproul [1] employed the bipolar pulse technique to reactively deposit films of TiO_2. They used a power supply, made by Magtron, which was capable of controlling the pulse parameters such as unipolar or bipolar mode, positive and negative pulse-on and pulse-off times, and frequency. They connected one side of the bipolar unit to the target and the other side to the chamber. The negative part of the cycle was on for 25 μs and off for 10 μs, whereas the positive part of the cycle was on for 5 μs and off for 10 μs. A reduction in the deposition rate, compared to that of the DC at the same power, was observed. It was attributed to the time of the positive part of the cycle as well as to the pulse-off time. Alternatively, Scholl [69] has designed a circuit containing an electronic switch, which may be controlled by a waveform generator, and a transformer. The output voltage of the transformer was approximately -0.1 of its input. The circuit periodically reverses the negative DC voltage, applied to the target, to a value approximately -0.1 of the operating target voltage. This voltage, which is slightly more positive than the plasma potential, allows electrons from the plasma to bombard the target and hence to discharge the insulating layer. The reversal time was approximately 5 μs, and the time between reversals was 50 μs. With such arrangements, Scholl [69] has reactively deposited highly insulating films, including Al_2O_3 and SiO_2.

Finally, a problem emerged during the non-reactive DC sputtering of carbon as an overcoat for computer disks. The origin of the problem was the formation of insulating islands, called 'nodules', on the carbon target which then could not be sputtered using the DC power. These nodules resulted from the implantation of hydrogen, formed by the disassociation of H_2O vapour presented in the vacuum chamber, into carbon. The application of low frequency AC power, as had been suggested by Cormia et al. [72], could solve the problem but the deposition rate was not satisfactory. This is because the deposition rate is proportional to the sputtering yield, which is in turn, proportional to the energy of the bombarding ions. Such an energy is proportional to the self-bias, and at the frequency suggested by Cormia, little negative self-bias can be achieved as the period was large compared with the transit time of the ions across the sheath. What is needed is a sufficiently small period, compared with the transit time of the ions across the sheath, so that ions do not have sufficient time to reach the cathode and neutralise the acquired negative charge. However, increasing the frequency leads to a significant increase in the deposition rate, but this solution is not cost effective since the cost of high-frequency AC power is higher than that of the DC power. To solve the problem, Scholl [86] suggested adding the AC (at 400 kHz) and the DC powers together through a 'combiner' which consisted of a high-pass filter in series with the AC power supply, and a low-pass filter in series with the DC power supply. High deposition rates were reported in the case of carbon. However, the deposition rate in the case of reactive sputtering of SiO_2 was quite low, and this was attributed to

the low levels of power that can be applied in this system and to the small size of the cathode compared with that of the substrate.

6.3.2.2. Bipolar pulsed magnetron sputtering using two magnetrons. To overcome the already mentioned drawbacks of Cormia's method, a sufficient number of electrons have to be made available to bombard the target when it is biased positively. In other words, an effective discharging of the insulating layer should be guaranteed for the sputtering process to be arc-free. On the other hand, the deposition of an insulating film also imposes another problem since the anode of the system will also be coated with the insulating material being deposited [1,69,74,76]. The anode, which may be the walls of the vacuum chamber or a separate electrode, acts as a collector of those electrons escaped from the plasma. The current they carry represents the return current to the power supply. Therefore, the increasing coverage of the anode with insulating layers, causes potential fluctuations and hence significant changes in the discharge parameters (i.e. an irretrievable drift in the desired set-point). Eventually, the discharge will extinguish due to the loss of an effective anode (i.e. the power supply comes to its voltage limit as a result of the increasing impedance). This problem becomes dominant in the industrial-scale production of optical films, such as SiO_2 and TiO_2. Therefore, the coating process has to be terminated periodically to allow the anode to be cleaned. Obviously, a well-defined DC conducting anode is required for an AC sputtering process to be stable at a given set-point.

The above requirements can be fulfilled by using a mid-frequency AC power supply to drive two magnetrons, which are decoupled from the sputtering chamber, side-by-side in a floating manner. The electrodes are then operated symmetrically. According to Fig. 13, in each half-cycle, one of magnetrons behaves as a temporary anode while the other behaves as a temporary cathode. The voltage waveform of the two electrodes are identical, provided that the material and the oxidation state of the two targets are also identical, but are always 180° out of phase regardless of the material and the oxidation state. This conclusion has also been supported by the work of Glocker [75] and Scherer et al. [77].

Fig. 14 shows the voltage waveform between one electrode and ground during the reactive deposition of AlN from two Al targets, using a 35-kHz AC power supply. The waveform of the other electrode, which does not appear in the figure, is identical but 180° out of phase [75]. If the area of the temporary anode is small, which is the case here, the potential across the anode sheath is positive with respect to the plasma potential, allowing more current to the anode. In fact, the potential of the temporary anode, in this technique, is usually of the order of only several tens of volts and

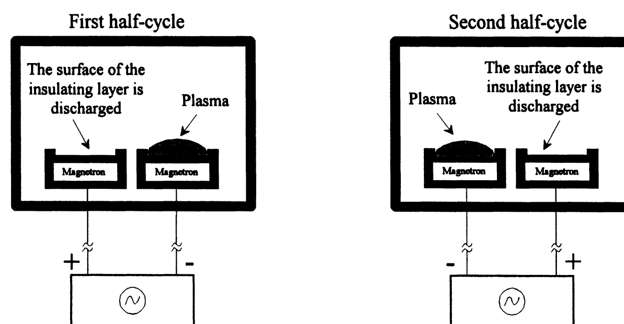

Fig. 13. Bipolar pulsed magnetron sputtering using two floating magnetrons.

is more positive than the positive plasma potential (usually of the order of several volts) [77]. Such a relatively small potential difference is sufficient for electrons, which are very mobile, to bombard the surface of the insulating layer to discharge it from the positive charge which had accumulated, during the negative half-cycle, as a result of bombardment by the very less mobile ions subjected to a relatively much higher negative potential difference. Thus, the requirement that, to avoid arcing ion and electron currents to the target must be the same, is fulfilled. Meanwhile, besides being sputtered, the temporary cathode provides the required electrons to discharge the surface of the insulating layer on the temporary anode. This periodical sputtering from each electrode results in a self-cleaning effect which leaves the centre of the erosion zone to be highly DC conducting which results in well-defined anodes and cathodes. Furthermore, such an arrangement allows for an effective decoupling of the processes on the electrodes from those on the substrate and the walls of the chamber. This is of great importance for the stability of a sputtering process in which the substrate is moving in front of the cathode [73].

Glocker [75] has studied the difference between 35-kHz AC and DC plasmas, during the reactive magnetron sputtering of AlN. His calorimetric measurements, at a total power of 500 W, revealed that the substrate temperature in the AC case was somewhat

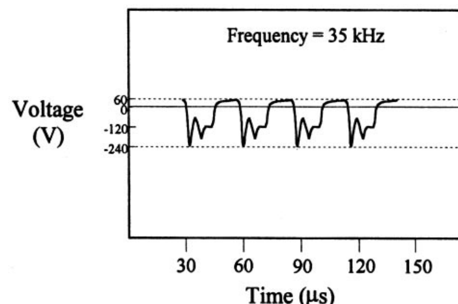

Fig. 14. Voltage between one cathode and ground during AC reactive sputtering of AlN. Reproduced from Schiller et al. [74].

higher than that of the DC. In terms of figures, he found that the average deposition rate and the energy delivered to the substrate per atom, in the DC case, were 0.82 nm/s and 20 eV, respectively, and 0.70 nm/s and 32.8 eV in the AC case. To find out the reason behind that, he used a Langmuir probe. His Langmuir probe measurements, under his experimental conditions, revealed:

1. the electron energies in the AC plasmas (3.2 eV) are slightly higher than in the DC plasmas (2.4 eV);
2. the differences between the plasma and the floating potentials are comparable (7 V in the AC case and 6.7 V in the DC case); and
3. the average ion densities in the AC plasma is approximately four times that of the DC plasma.

The last difference, between the AC and the DC plasmas, is the most significant. According to optical emission measurements, the plasma extinguishes on each half-cycle and has to be reignited. The increase in ion densities in the AC plasma was attributed to target voltage spikes during the reignition of the plasma on each half-cycle, as it is evident on the negative-going part of each cycle (Fig. 14). Such spikes cause rapid electron acceleration in the presheath region leading to significantly more efficient ionisation of gas and hence much higher plasma bombardment.

This emerging technique has been utilised recently by a few workers [73–75,77,82]. The results were very promising. For example, Szczyrbowski and Braatz [73] have reactively deposited films of SiO_2 at high rates using 40-kHz AC power applied between two Si magnetrons. In addition to the excellent optical and mechanical properties of the deposited films, no arcing was observed during the entire lifetime of the target which was more than 1 week. Schiller et al. [74] have reactively deposited films of Al_2O_3 from two Al magnetrons using an AC power at different frequencies ranged from 50 Hz to 164 kHz. They reported a significant decrease in the defect density of the deposited films with increasing frequency. The curve, for non-absorbing films, saturated at frequencies greater than 50 kHz which was an indication of an arc-free process beyond this frequency. They also reported a deposition rate of approximately 60% of that of metallic Al. The rate was almost independent of the frequency in the range they investigated. Scherer et al. [77] have also adopted the two cathodes technique to reactively deposit films of Al_2O_3, SiO_2 and Si_3N_4 using 40-kHz AC power. They reported deposition rates comparable to those obtained with the DC power.

To summarise, bipolar pulsed magnetron sputtering technique, using two magnetrons, enjoys the following advantages:

1. The periodical effective discharging of the insulating layer, due to the symmetrical operation of the electrodes, allows the reactive sputtering process to be arc-free [77]. Consequently, the defect density in insulating films is reduced by orders of magnitude [74] in comparison with the DC technique.
2. The well-defined DC conducting anode allows the sputtering process to be stable at a given set-point.
3. The high deposition rates obtained are comparable with those of the DC technique [69,73,77].
4. Unlike the additional complexity of the RF technique, the coupling of the AC power to the cathodes, in the frequency range used, is simple. Consequently, the AC technique can be easily adopted for sputtering from larger area cathodes [77].
5. It is important to note that the deposition of highly insulating films creates another problem regarding ion-bombardment of the growing film. The accumulation of positive charge on the surface of the film prevents any further bombardment. The AC technique helps in discharging the surface of the growing film during the positive half-cycle, allowing continuous bombardment [1].
6. The substrate temperature in AC plasmas is higher than that in similar DC plasmas [75].

7. Conclusion

In reactive sputtering, the presence of the reactive gas, at both the cathode surface and the substrate, results in strong interactions of the reactive gas not only with the condensing material at the substrate but also with the cathode surface, the so-called target poisoning. If flow control of reactive gas is used, the very undesirable hysteresis effect occurs and the process becomes unstable.

There are several expensive and/or impractical methods to stabilise the process, such as

1. increasing the pumping speed to be greater than $-\mathrm{d}f_r^{sm}/\mathrm{d}P_r$: the problem with this method, especially in industry where large vacuum chambers may be used, lies in adding considerable costs at the building stage of new machines, and can be difficult to adopt in existing ones;
2. increasing the target-to-substrate distance, which requires larger vacuum chambers, and hence, higher costs; it also results in lower deposition rates;
3. obstructing reactive gas flow to the cathode, which

needs complicated arrangements and results in lower deposition rates; and
4. pulsed reactive gas flow, which needs an extensive amount of process optimisation prior to the reproducible deposition of films with the required stoichiometry — it also requires a continuous monitoring and adjustment of the process parameters.

Plasma emission monitoring and voltage control are two inexpensive and practical methods of control to prevent instability. They have proved to be powerful techniques for monitoring and controlling the reactive sputtering processes for the reproducible deposition of high-quality films. They sample from the region where the reaction is actually taking place, the information may be obtained in real time without disturbing the discharge. The drawbacks of plasma emission monitoring, such as the sensitivity of the signal to the sampling position relative to target and the change of the signal with target erosion, are insignificant. They can be overcome when the relative values of the signal, in a set of experiments, are considered. In reality, the absolute value of the PEM signal has no practical importance. On the other hand, voltage control may be employed in certain conditions. The difference between the cathode voltage of the metallic target material and that of the reaction product should be large and positive. Furthermore, the cathode voltage, as a function of the reactive gas flow rate, or its partial pressure, should be single valued.

The serious problem encountered during the reactive deposition of highly insulating films from metallic targets, using DC magnetron sputtering technique, is the build-up of an electrically insulating layer on the cathode surface with the resultant charge accumulation and arcing. The latter may be initiated by the breakdown of an insulating layer which builds up outside the racetrack of a magnetron cathode. Arcs have very destructive effects, such as driving the process to become unstable, reducing the target lifetime and creating defects in the sputtered films.

To secure an efficient avoidance of arc formation, without affecting the deposition rate, the discharging frequencies of the insulating layers should be between approximately 10 and 70 kHz. Unipolar or bipolar pulsing techniques, using one or two magnetrons, may be employed.

Bipolar pulsed magnetron sputtering technique, using two magnetrons, enjoys many advantages. In addition to high deposition rates, the simplicity of coupling the AC power to the cathode, the continuous bombardment of highly insulating films and the capability of sputtering from large area cathodes at higher substrate temperature than that in similar DC plasmas, bipolar pulsed magnetron sputtering technique allows the process to be stable and arc-free with the resultant significant reduction of the defect density in insulating films.

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