

# Physical investigation of copper thin film prepared by sputtering

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Copper thin films have been deposited by sputtering. The simulation results show that most of deposited atoms on the substrate have kinetic energy ranging between 1 and 1.5 eV which leads to the deterioration of the crystalline quality, the impact angle between  $10^\circ$ -  $70^\circ$  and  $100^\circ$ - $160^\circ$ , with a peak at  $40^\circ$  and  $150^\circ$ , the oblique incidences of sputtered atoms on the substrate give films with porous and columnar microstructures. For higher pressure these distributions are more uniform leading to the uniformity of the obtained thin film which paid a great attention in the industry. These results are in accordance with numerical and experimental ones obtained in others works.

**Keyword:** Monte Carlo simulation, Growth, Sputtered atom, Thin film

## 1 Introduction

Copper is an attractive interconnecting material for current due to its low resistivity and good electromigration resistance<sup>1-3</sup>. Thin films are widely used in many fields such as optics, microelectronics, solar energy, biomedical, magnetic materials and so on<sup>4-9</sup>. Copper thin films are deposited by various techniques such as evaporation, sputtering, chemical vapor deposition, electroplating, and ion beam deposition<sup>10-16</sup>. Among these techniques, sputtering deposition is one of the best methods, for practical preparation of thin films. It has been shown that sputtering offers more flexibility to the process and gives good quality of films with repeatable performance<sup>17-19</sup>. Homogeneity of the film thickness is a basic demand in thin film technology. Many factors govern the characteristics of the deposited films, especially the pressure of the background gas<sup>20,21</sup> and consequently the impact angle and the energy of deposited atoms on the substrate. Therefore we have to find a relationship between pressure, impact angle, energy and uniformity of the film. However, the impact angle and the energy are difficult to measure or calculate. Therefore, the use of computer simulations can be effective. For that we have developed a numerical model based on the results of the collisional cascade sputtering theory to simulate the transport process, introduced by Motohiro and Taga<sup>22-24</sup> and Somekh<sup>25</sup>. The initial energy distribution is well described by the Sigmund-

Thompson<sup>26</sup>, this distribution is based on the linear cascade theory. The angular distribution of the sputtered atoms is usually considered to be  $\cos^n \theta$  type depending on the energy and the angle of incidence of the impinging atoms. This Monte Carlo method can provide various kinds of information on the properties of sputtered particles flows being deposited on the substrate, and gives considerable details of the sputtered processes. This procedure makes possible to obtain spatial, energetic and angular distribution of sputtered atoms on the substrate surface.

In this study, the transport process of sputter atoms from a Cu target to a substrate through Argon plasma in direct current sputtering (D.C) deposition was investigated by Monte Carlo simulation. Deposition profiles have been obtained at different processing condition.

## 2 Model and Method

The Monte Carlo model used in the present study was developed earlier by several groups<sup>22,23,25</sup>. This approach is based on few assumptions to simplify the complexity involved in the sputtering process of Cu atoms. These assumptions are as follows:

- (i) The concentration of the sputtered species is small when compared with that of the background gas.
- (ii) Ions impinge normally to the target.
- (iii) The kinetic energy of the ions depends to the target voltage.
- (iv) The angular and energy distributions of the

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ejected species obey a theoretical expression derived by Sigmund-Thompson<sup>26</sup>.

- (v) The mutual collisions between the sputtered atoms are neglected.
- (vi) The collisions between the sputtered atoms with the background gas atoms are elastic.
- (vii) The background gas pressure is assumed to be spatially uniform.
- (viii) The sticking coefficient is set to unity at the substrate surface.

These assumptions have been justified in earlier reports<sup>22,25,27,28</sup>.

The distribution of the initial energy,  $E_0$ , and initial angle,  $\theta_0$ , of the sputtered atoms leaving the cathode is given by Sigmund-Thompson's formula<sup>26</sup> as:

$$\psi(E_0, \alpha_0) = \frac{2UE_0}{(E_0 + U)^3} \frac{\cos \alpha_0}{\pi}, \quad \dots (1)$$

where  $U$  is the surface binding energy of the cathode material, taken to be the sublimation energy ( $U = 3.49$  eV). Using random number<sup>29</sup> between 0 and 1, the angle and energy of a sputtered atom starting at the cathode are calculated from Eq. (1) as:

$$\alpha_0 = \left(\frac{1}{2}\right)a \cos(1 - 2h_1), \quad \dots (2)$$

$$E_0^2(h_2 - 1) + E(2Uh_2) + U^2h_2 = 0, \quad \dots (3)$$

The azimuthal angle  $\beta$  (the angle of the projection of the velocity vector in the  $xy$  plane, with respect to the  $x$  axis, see Fig. 1) is also generated uniformly between 0 and  $2\pi$  by a random number  $h_3$  between 0 and 1:

$$\beta = 2\pi h_3 \quad \dots (4)$$

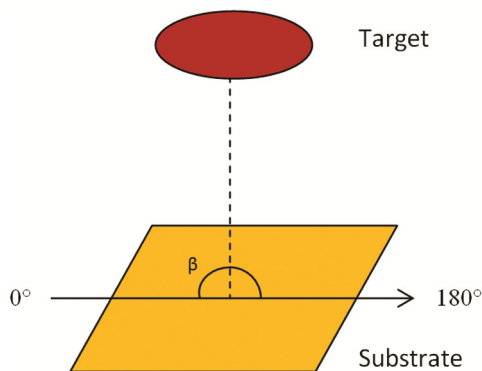


Fig. 1 — Model illustration used in the simulations.

The  $x$  and  $y$  coordinates of the starting position are also determined by random number  $h_4$ :

$$x = \frac{D}{2} h_4 \sin \beta, \quad \dots (5)$$

$$y = \frac{D}{2} h_4 \cos \beta, \quad \dots (6)$$

The starting  $z$  coordinate is zero. The target diameter ( $D$ ) has been used as 40 mm.

Calculating the gas phase transport processes of an ejected particle, the Monte Carlo method<sup>22,23</sup> was used. Because of the high kinetic energy of sputtered particles in comparison with the ambient gas atoms, the motion of gas atoms will be neglected. Therefore the mean free path  $\lambda_m$  of a sputtered particle is obtain by:

$$\lambda_m = \frac{k_b T}{r_{sg}^2 \pi p}, \quad \dots (7)$$

where  $k_b$  is Boltzmann constant,  $T$  is the temperature of the sputtered gas taken as 350 K,  $p$  is the pressure of the sputtered gas, and  $r_{sg}$  is the inter-atomic separation of sputtered particle and working gas atom given by:

$$r_{sg} = r_s + r_g \approx \frac{-2}{b_s + b_g} \ln \frac{E_s}{\sqrt{A_s A_g}}, \quad \dots (8)$$

where  $r_s$  and  $r_g$  are the atomic radius of the separation of sputtered particle and working gas,  $E_s$  is the kinetic energy of the sputtered atom and  $A_s$ ,  $b_s$ ,  $A_g$  and  $b_g$  are Borne-Mayer parameters for the sputtered and the background gas atoms respectively<sup>30</sup>.

During the gas phase transport, the collisions between the sputtered particle and the background gas are assumed to be a random process described by a Poisson distribution. Therefore, the distance between two successive collisions  $\lambda$  is generally determined from the Poisson distribution according to a standard M.C procedure;

$$\lambda = -\lambda_m \ln(h_5), \quad \dots (9)$$

Where  $h_5$  is a random number.

The interaction between atoms is described by an interaction potential, which can be either attractive

potential or repulsive potential. One of widely used repulsive potential is the Borne-Mayer potential<sup>30</sup>. After a collision between the sputtered atom and the sputtered gas atom, the sputtered particle is scattered and thus change its direction and energy. According to the hard sphere collision model which is used to study the trajectory of ejected particle, the scattering angle  $\chi$ , azimuthal angle of scattering  $\psi$  and energy loss rate  $\alpha$  in the laboratory are given by:

$$\tan \chi = \frac{\sin(\pi - 2\alpha)}{(m_1/m_2) + \cos(\pi - 2\alpha)}, \quad \dots (10)$$

$$\psi = 2\pi h_6, \quad \dots (11)$$

$$\eta = \frac{(m_1 - m_2)^2 \cos^2 \alpha + (m_1 + m_2)^2 \sin^2 \alpha}{(m_1 + m_2)}, \quad \dots (12)$$

Where  $m_1$  and  $m_2$  are the mass of the sputtered and background gas atoms respectively,  $\alpha$  is angle between the line which passes through the centers of the sputtered atom and gas atom on collision.  $h_6$  is the random number. The direction of velocity of the sputtered atom before collision is calculated using random number  $h_7$ :

$$\alpha = \arcsin(h_7)^{1/2}. \quad \dots (13)$$

The new direction in three dimensional, i.e., the angle  $\alpha$  and  $\beta$  are calculated from the angle  $\alpha_0$  and  $\beta_0$  before scattering and from the scattering angle  $\chi$  and azimuthal angle of scattering<sup>20</sup>  $\psi$ . The new energy  $E_1$  and the new position ( $X_1, Y_1, Z_1$ ) of the particle after collision in laboratory system can be calculated from the elementary process of hard sphere collision as :

$$E_1 = \eta \cdot E, \quad \dots (14)$$

$$\begin{pmatrix} X_1 \\ Y_1 \\ Z_1 \end{pmatrix} = \begin{pmatrix} X_0 \\ Y_0 \\ Z_0 \end{pmatrix} + \begin{pmatrix} \sin(\alpha)\cos(\beta) \\ \sin(\alpha)\sin(\beta) \\ \cos(\beta) \end{pmatrix} \lambda. \quad \dots (15)$$

After the new energy, position and direction are determined, the atom is followed during the next step, and the process is repeated. The atoms are followed until they have thermal energies ( $E < 0.038$  eV for

$T=350$  K). Similar trajectories have been simulated for  $10^5$  atoms. In this way the characteristic of sputters atoms are obtained.

### 3 Results and Discussion

#### 3.1 Transport efficiency

In all cases, a substrate of  $60 \times 60$  mm was considered, and was placed on plane parallel to the surface of the target at the distance of 25 mm. One important result of sputter atom transport simulation using the Monte Carlo model is the deposition efficiency, which depends strongly on the gas pressure, the distance between the target and the substrate, the collision probability, the atomic number, the mass ratio of the sputtered atoms, the gas atoms and other factors<sup>31-36</sup>. Figure 2 shows the simulation result of the sputtered copper atoms transported and impinging at the substrate in the pressure range of 1 – 20 Pa. It can be seen, that a reduction of the flux is observed for copper, with the increase in the sputtering pressure. The factor responsible for this phenomenon is the thermalization of sputtered atoms. Before the deposition of copper atom, a sputtered atom undergoes a lot of collisions with the background gas atom; and every collision induces the energy variation. At lower pressure, collision of sputtered particles with the background gas is not the dominant process, thus the number of thermalized atoms diminishes, and hence the amount of sputtered atoms striking the substrate increases. However, at higher pressure the mean free path of sputtered atoms is very short than they undergoes more collisions and become fully thermalized with the background gas, and they can't reach the substrate. As

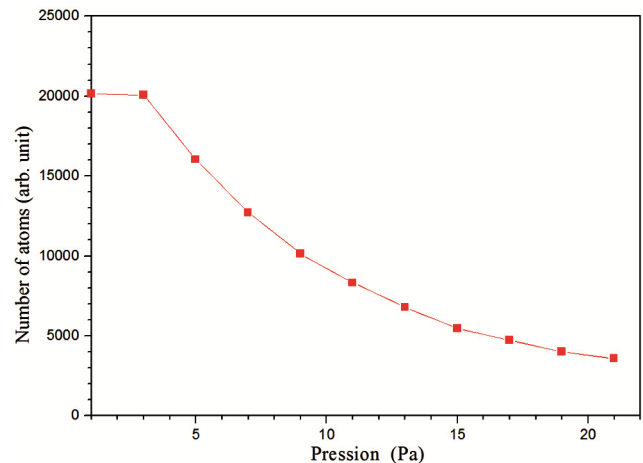


Fig. 2 — Number of atoms reaching the substrate as a function of the sputtering pressure.

consequence, the amount of sputtered particles reaching the substrate diminishes very quickly. This result is in accordance with the numerical and experimental results reported in<sup>21,32,36,37</sup>. It is obvious that the energy and the direction of these particles arriving at the substrate are in close relation with the transport process. The code can be used to calculate the impact angle and energy of sputtered particles arriving at the substrate, which are both important parameters<sup>38</sup>.

### 3.2 Impact angle and energy distribution

The dependence of incident angle of sputtered atoms deposited on the substrate in the sputtering pressure range of 1 - 20 Pa has been evaluated and shown in Fig. 3. At all pressures, the distribution exhibits a broader impact angle range between 10°-70° and 100°-160° with a peak at 40°, and no significant deviations of sputtered atoms are observed. For lower pressure range, the sputtered atoms preserve the cosines law distribution, due to small numbers of scattering. For higher pressure range, the number of collisions of atoms increases, than scattering of sputtered particles also increases. Similar behaviour has been observed by several researchers<sup>36,38</sup>. The oblique incidences of sputtered atoms on the substrate give films with porous and columnar microstructures<sup>36</sup>. Finally, the difference of the angular distribution for various pressures will be analyzed because it has a significant impact on the film growth characteristic<sup>36,39</sup>. The film is also affected by the incident energy, so we must also simulate the effect of the pressure on the average energy of arriving atoms.

Figure 4 shows the simulated incident energy of Cu specie impinging at the substrate for different sputtering pressures. We can observe, that whatever the pressure, most of the deposited atoms have their energy less than 1.5 eV. The higher energy exceeds 2 eV but with very small number of atoms. As consequences, the copper atoms arrive on the substrate after higher number of collisions, i.e., with lower energy. When the atoms reach the substrate surface, they have not enough energy and consequently the sputtered atoms have low mobility and the formation of crystalline lattice will be more irregular and leads to the deterioration of the crystalline quality<sup>40</sup>. At lower pressure the distribution of the incident energy is non uniform with an increase in low energy content. At higher pressure, the energy distribution becomes more uniform, which leads to

structural uniformity<sup>36</sup> but with very few atoms because of the increase of the collisions number. The optimization of the sputtering pressures gives sufficient and homogeneous energy distribution. The above results are significant since the properties of the thin film, stress, microstructure and surface roughness are observed to be dependent on energetic bombardment<sup>20</sup>. The simulated results by Monte Carlo technique are in accordance with the results reported in<sup>36,41</sup>. Our code can also be used to simulate the thickness profile, which is important parameter to test the final film quality.

### 3.3 Thickness profile

Figure 5(a-c) shows the representative plot of the influence of sputtering pressure on the simulated 3D spatial profiles of Cu incident atoms reaching the substrate. At low pressure, there are a large number of atoms deposited on the substrate, this is due to the reduction of the collisions rate of the atoms of the material with the gas atoms, but the film obtained is not uniform. By increasing the pressure, the collision

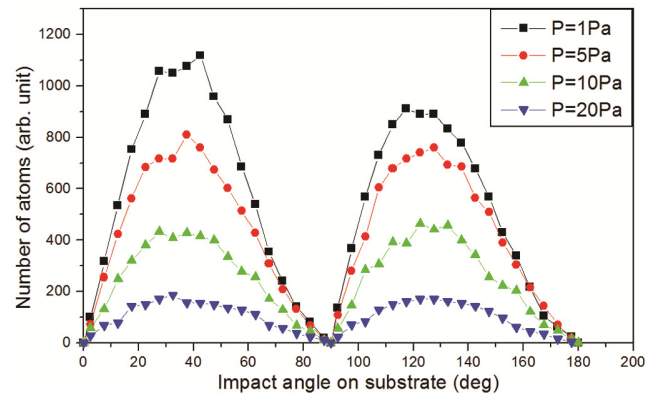


Fig. 3 — Impact angular distribution of sputtered atoms on the substrate for various pressures.

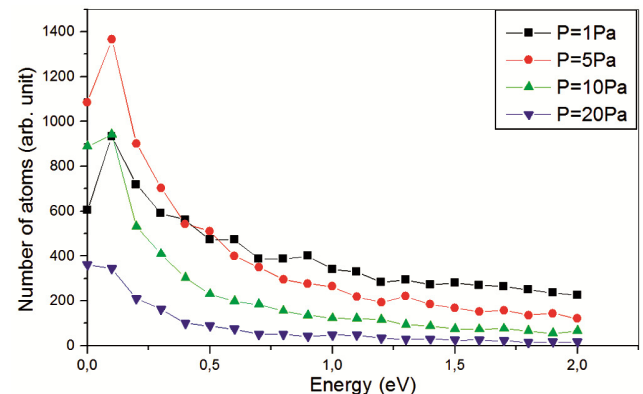


Fig. 4 — Kinetic energy of sputtered Cu atoms incident on substrate for various sputtering pressures.

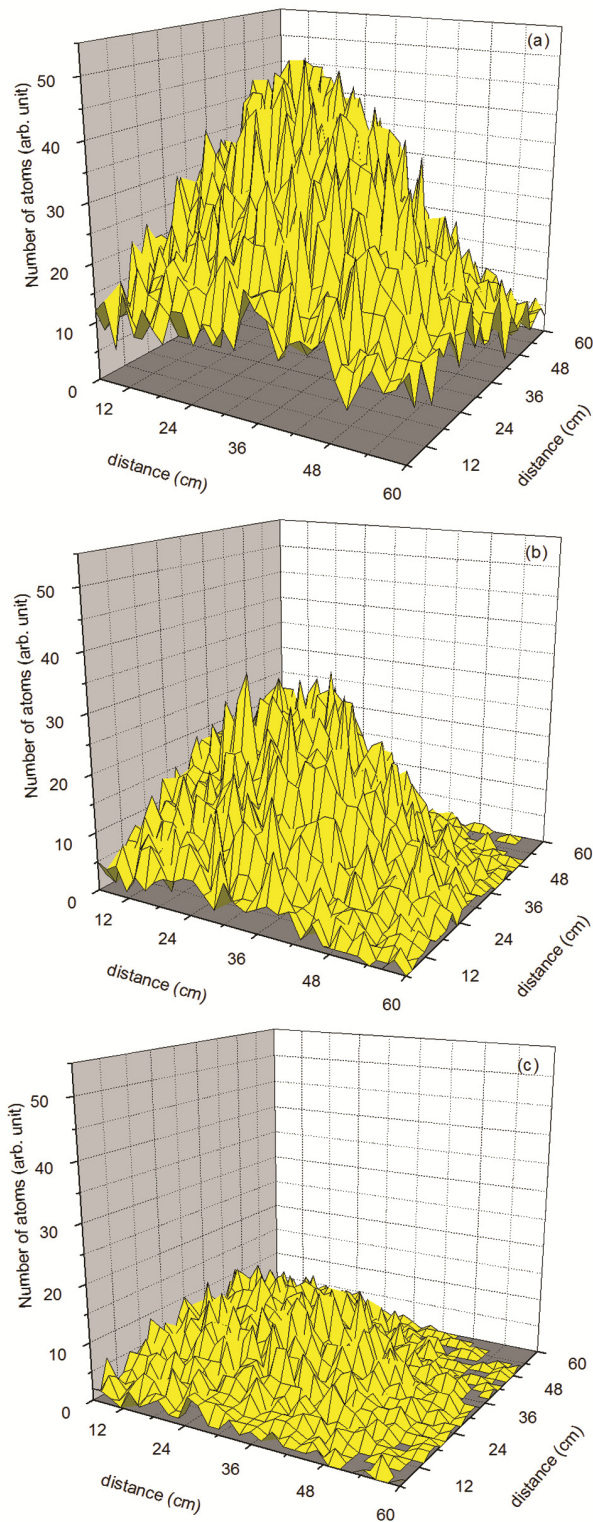


Fig. 5 — 3D spatial distribution of the sputtered flux on the substrate at (a)  $P=1$  Pa, (b)  $=10$  Pa and (c)  $=20$  Pa.

rate increases, most atoms lose their energy and therefore cannot reach the substrate, but the resulting

film is more uniform compared to the low pressure, thanks to the phenomenon of collision<sup>36</sup>. It was observed experimentally by Mishara *et al.*<sup>42</sup> that the deposited films at higher pressure had larger particle sizes, therefore the electrical resistivity is also affected<sup>40</sup> knowing that copper films layers are used in interconnecting materials. Consequently the pressure has a significant effect on the film growth. The above observation was also confirmed experimentally by Nathan *et al.*<sup>43</sup> and Myer *et al.*<sup>44</sup> and numerically by Nimisha *et al.*<sup>36</sup> and Castro *et al.*<sup>40</sup>. The obtained results could be found to be useful in understanding the effect of process parameters on transport and consequently in understanding the sputter deposition process.

#### 4 Conclusions

We have simulated by Monte Carlo method the transport of copper atoms using D.C sputtering configuration in argon gas at various sputtering pressure for constant distance target-substrate. A collisionnel model developed by Born- Mayer was used to achieve the simulation. The obtained film depends strongly on the gas pressure and consequently on the angular and the energetic distribution. By decreasing the pressure, the atoms deposition increases but the uniformity of the film will be reduced. While increasing the pressure, the deposit decreases but the obtained layer is more uniform, therefore the deposited film must be optimized. Currently the challenge for researchers is focused on the uniformity and electrical characteristic of layers; it is thus necessary to consider the energetic and the angular distribution of sputtered atoms. By comparing our results with others experimental and theoretical works, it can be noted that our generated code is valid and can be applied to other novel deposition processes. Finally, we believe, that Monte Carlo simulation provide a new window of process conditions for the sputtering deposition.

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