The Growth of Single Crystal Films of Silver on Rocksalt by Sputtering

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Thin films of silver on rocksalt have been prepared by diode sputtering in an argon discharge. Deposition rate, substrate temperature, and film thickness have been varied, and dependence of orientation on these parameters has been studied. Within a deposition rate range of 0.1 to 1.15\AA/sec , films have been grown with (100) $[110]_{A_\text{g}}$ / (100) $[110]_{\text{NaCl}}$ orientation, at temperatures in the range -35 to 0° C. Higher rates required higher temperatures. Films giving these results were all 200 \AA in thickness. A thickness dependence of orientation has been observed for films below 120 A in thickness.

The above results are discussed in terms of the effect of arrival energy of the sputtered material at the substrate. The results of calculations, on the effect of gas pressure on the arrival energy, are presented and it is shown that, at 10^{-2} torr, up to 15 $\%$ of the arriving atoms will have energies above 0.6 eV. That the observed rate, temperature, and thickness dependence of epitaxy are due to surface cleaning and penetration effects caused by the energy of arrival of the atoms is shown to be possible.

The effect of charged particle bombardment of the substrate is also considered. It is shown that this may also be an important parameter affecting the growth.

I. Introduction

The preparation of thin films by sputtering has become of increasing interest recently, since it has been shown possible to grow, thereby, epitaxial films at lower substrate temperatures than by evaporation [1]. Even for similar substrate temperatures, differences in properties are found to exist; Chopra *et al* [2, 3] have studied this with respect to gold using electron microscopy and diffraction and by the examination of electrical properties. They found that sputtered films approach bulk resistivity at a lower thickness than evaporated ones do, and they explain this in terms of island size; evaporated films having smaller, more numerous islands in the initial stages of growth. There is a consequent increase in the defect density in the continuous film, and the final resistivity of an evaporated film is higher than that of a sputtered one. Similar work has been undertaken with silver [4].

The purpose of the present paper is to examine

in detail the parameters influencing epitaxia growth of sputtered silver films on rocksalt. The effect of rate of deposition, substrate temperature, and film thickness has been investigated.

2. Experimental Apparatus

The vacuum apparatus used was a 12in. diameter \times 14 in. high, glass chamber evacuated by a 4 in., oil diffusion pump backed by an oil rotary pump $(1 \text{ in.} = 2.5 \text{ cm})$. With liquidnitrogen trapping, an ultimate pressure of 1.10^{-6} torr was obtainable. The ht voltage for sputtering was obtained unsmoothed through a bridge rectifier from a 5 kV, 50 mA transformer.

The mode of sputtering was of the simple diode type, in which the material to be sputtered is made the cathode in a parallel planar electrode arrangement. A negative potential was applied to the cathode and the substrate was placed on the anode, which was at earth potential. The electrode separation was maintained at 10 cm, except for the low-temperature experiments, in

which the separation was 8 cm. The electrode arrangement was such that the discharge always took place between cathode and anode, other earth points being further away from the cathode or within the discharge dark space.

The pressure of the gas for diode sputtering experiments in a glow discharge is approximately $1.\overline{10^{-2}}$ torr, but in all cases the system was evacuated to $\langle 2.10^{-5}$ torr prior to introduction of the discharge gas. Argon of 99.999 $\%$ purity was used as the discharge gas and was supplied to the system from a cylinder, passing through a drying agent to a reservoir kept at a constant, positive, argon pressure. No special measures were taken to further purify the argon.

Substrates were prepared by cleaving NaC1 in air, to reveal a (I 00) face, immediately prior to placing in the vacuum system.

For a 3 kV applied potential, the discharge current was varied over the range 2 to 200 μ A/ cm 2 by means of adjustment of the gas pressure; higher pressures gave higher currents by virtue of the increased ionisation. The rate of ejection of atoms from the target, and hence the rate of deposition onto the substrate, is proportional to the bombarding ion density; i.e. the ion current density. The above current range corresponded to a deposition rate range of 0.02 to 2.0 A/sec.

The final, average film thickness was measured by means of a surface probe across a scratch made through the film deposited on a glass monitor slide $[5]$. Films less than 100 Å thick were estimated by extrapolation from a thickness-time plot. Such an extrapolation assumed a constant condensation coefficient during growth.

The substrate temperature was measured by means of a fine-wire, Chromel/Alumel or Pt/Pt- 13% Rh thermocouple, placed in contact with the substrate surface. In order to obtain reproducible low temperatures, it was necessary to use a totally enclosed sputtering assembly. It was found that simply placing the substrate on a fiat plate cooled to liquid-nitrogen temperature was unsatisfactory. Even if the cooled plate was at -196 ° C, the surface of the rocksalt substrate was only approximately -50° C. Clamping the substrate to the plate over the whole substrate area produced a lower temperature, but this was impracticable experimentally. Floating the specimen on a mercury pool, which was then frozen, enabled -120° C to be reached, but in such cases mercury contamination of the substrate was unavoidable.

The sputtering module used for the low temperature depositions was similar to that described by Theurer and Hauser [6], in which the cathode and substrate support were totally enclosed in a liquid-nitrogen-cooled, stainlesssteel container. The discharge occurred between the cathode and the walls of the container. The substrate support was a tantalum strip through which a current could be passed to increase the substrate temperature. The support was situated outside the dark space of the discharge. Temperatures in the range -60° C to room temperature could be readily attained by controlling the liquid-nitrogen flow and the heater current.

Temperatures above room temperature were realised by allowing the discharge to heat the substrate to equilibrium temperature prior to deposition.

In the thickness-dependence experiments, it was necessary to have a shutter system, whereby successive substrates could be exposed to the sputtered material.

For films less than \sim 100 Å in thickness, an overlayer of platinum-carbon was evaporated onto the substrates to support the films for electron microscope examination. The evaporation was carried out immediately after sputtering, the system being first evacuated from 10^{-2} to 5.10^{-5} torr. Stirland and Campbell [7] have shown that, if the system is let up to atmospheric pressure between the metal film deposition and the carbon evaporation, then the metal deposit may move around on the rocksalt surface.

Specimens were examined by conventional electron microscopy, in a Siemens Elmiskop I operating at 80 or 100 kV. The films were supported on a 200-mesh grid, after floating off the substrate in deionised water.

3. Results

3.1. Variation of Crystal Structure with Rate and Temperature of Deposition

To study the effect of rate and temperature of deposition, films were prepared at rates ranging from 0.02 to 1.25 Å/sec and temperatures ranging from -55 to $+55$ °C. Film thickness in all cases was 150 to 200 A.

Results showed that high deposition rates required a correspondingly higher substrate temperature for single crystal growth than films prepared at a lower rate. The results are shown graphically in fig. 1, in which the deposition rate is plotted as a log function against the reciprocal of the absolute temperature. Such a plot is

Figure 1 The **orientation dependence** of sputtered silver on rocksalt as a function of rate of deposition and substrate temperature (film thickness 200 Å).

derived from an energy equation of the type $R = R_0 \exp(-E/kT)$. On this plot, a boundary line can be drawn, on one side of which the films are single crystal, and on the other side of which they are polycrystalline. Films were deemed to be single crystal or polycrystalline according to their diffraction patterns, and three typical patterns are shown on the figure. Distinction near the boundary was more difficult, but any streaking of the spots or the presence of any (111) rings was taken to indicate that the film was polycrystalline.

The orientation of the single crystals was always observed to be (100) $[110]_{Ag}$ // (100) $[110]$ _{NaCl} and very little trace of (111) $[110]$ _{Ag} // (100) [110] $_{\text{NaCl}}$. A large number of structural faults was evident and some of these showed up as twin and double diffraction spots in the diffraction patterns.

3.2. Variation of Crystal Structure with Thickness

Using a rate and temperature known to give single crystals at thicknesses of 200 A, films were prepared with average thicknesses of 4 A upwards.

Initial experiments were carried out at a rate of 0.25 A/sec and a substrate temperature of 25° C. The films were backed with platinumcarbon, as described earlier. The films so prepared were found to be made up of discrete islands as expected, but the diffraction pattern from these films showed a distinct increase in film orientation as the thickness increased.

In order to ensure that this result was not due to the small islands moving or detaching in the stripping process, the experiment was repeated using a deposition rate of 0.12 Å/sec, and an overlayer of \sim 150 Å of nickel was evaporated

onto the silver films immediately after these had been deposited. The nickel deposition rates were fairly high (\sim 30 Å/sec) and the pressure prior to evaporation was 1.10^{-5} torr.

The results of these experiments again showed a marked orientation dependence upon thickness. A sequence of films at different thicknesses is shown in fig. 2a to c. The major pattern is due to nickel, but the ring marked (220) is that for silver. At 7 Å , the silver is polycrystalline, the degree of orientation improving with thickness, becoming virtually single crystal at 115 A. A diffraction pattern from a 200 A film, grown under identical conditions but without a nickel-backing film, is shown in fig. 2d.

4. Special Features of Sputtering Deposition

4.1. Introduction

Comparison of films grown by evaporation and by sputtering shows that single crystal films can be produced by the latter method when the substrate temperature is much less than that required for the evaporation method [8]. Two major differences exist between the two methods. The energy of atoms leaving the target is at least an order of magnitude greater than that of evaporated ones [9]. Also, there is the possible existence of negative ion or electron bombardment of substrates on the anode, which would serve to modify the substrate surface during deposition.

4.2. Arrival Energy of Atoms at the Substrate

Owing to the presence of gas in the interelectrode region, sputtered particles leaving the target make collisions with gas atoms and ions on their way to the substrate. The number of collisions will depend upon the gas pressure, and the energy lost will be dependent upon the masses of the colliding particles.

The problem is to find how the initial energy distribution of sputtered particles changes as a function of gas pressure and interelectrode spacing. The following approach is a relatively simple one and is based on average effects rather than single effects. For ease of computation, velocity rather than energy distributions have been considered.

For the results to be of interest, we require to know what fraction of the sputtered distribution arrives at the substrate with an energy significantly greater than that of thermally evaporated atoms. For atoms having comparable 370

energies to thermally evaporated atoms, the deposition will be fundamentally the same as it is in evaporation. An energy distribution for silver evaporated from an open source at 1800° K will be Maxwellian, with a most probable velocity of \sim 3 \times 10⁴ cm/sec. Experimental evidence for Maxwellian distributions from evaporation sources has been given for potassium and thallium by Miller and Kusch [10]. A very small number will have velocities above 1×10^5 cm/ sec, and the calculations have thus been concerned with establishing the numbers of sputtered atoms that arrive at the substrate with velocities above this value.

The sputtered particles are ejected in all directions from a polycrystalline target and a nearcosine distribution exists [11]. For ease of calculation, however, only those travelling normally from the target have been considered, and it is assumed that all colliding target atoms continue in a forward direction following collision. Velocity distributions for ejection normally from a polycrystalline target have been given by Stuart and Wehner [9], for the case of copper bombarded with mercury ions at energies up to 600 eV. Such a curve is approximately Maxwellian, but with a high-energy tail; the peak velocity is 3.5×10^5 cm/sec (mean velocity \sim 4 \times 10⁵ cm/sec). In the absence of velocity data for silver bombarded with argon at 3 keV, this distribution in velocity has been used. Certainly there appears to be little change in peak velocity with bombarding energies up to 600 eV [9], and it is probable that the peak velocity will therefore not vary appreciably up to 3 keV. However, the high-energy tail will almost certainly be lengthened. Very high-energy (45 keV) bombardment of polycrystalline silver with argon ions has been considered by Almén and Bruce [12], and they have shown that the mean velocity in the direction normal to the target is 7.1×10^5 cm/sec. There is thus no very large rise in the peak velocity with bombarding energies, and it is therefore felt that the use of the Wehner velocity distribution can give useful information which will not be too far from the actual distribution behaviour.

Under the conditions of our experiments, the ion current density is of the order of 25 μ A/cm², which corresponds to an ion impingement rate of \sim 10¹⁴ ions/cm² sec. For a gas at 300° K, the number of atoms striking unit area per sec at 1×10^{-2} torr is $\sim 10^{18}$, so the ratio of atoms to ions is $\sim 10^{4}$:1. Collisions between sputtered

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 (a)

 (b)

Figure 2 Electron diffraction patterns showing change of orientation with thickness (rate of deposition 0.12 Å/sec; substrate temperature 25° C): (a) 7 A thick, nickel backed; (b) 25 A thick, nickel backed; (c) 115 A thick, nickel backed; (d) 220 Å thick, no backing.

particles and discharge gas ions can therefore be neglected.

To simplify the calculation, the gas is assumed to be stationary. This is a reasonable assumption since, for a Maxwellian distribution, only 0.01 $\%$ of the molecules have $v > 3\bar{v}$, where \bar{v} is the mean velocity of the distribution. For argon at 300° K, $\bar{v} = 4 \times 10^4$ cm/sec, while, as we have discussed, the sputtered particles are considered to have $\bar{\nu}$ around 4×10^5 cm/sec. If the sputtered particles have velocities approaching that of the gas atoms, then the stationary gas assumption breaks down. However, we have already decided to ignore sputtered atoms with velocities below 1×10^5 cm/sec, so that the stationary gas approximation will be satisfactory.

Let the space between the electrodes be made up of n slabs, of unit cross-sectional area and thickness δx . If there are N atoms per unit volume, then each slab will contain $N\delta x$ atoms, which will present an area $N\delta xq$ to the impingeing beam, provided δx is small enough to contain only a negligible fraction of atoms lying behind one another; q is the collision cross-section. In general, for n slabs, the area A presented to the beam is given by

$$
A=1-(1-p)^n
$$

where $p = N\delta xq$.

The fraction of incident atoms which get through *n* slabs without collisions is $(1 - A)$. The error in $(1 - A)$ depends upon the number of slabs chosen. For an interelectrode separation of 10 cm, divided into 100 slabs of 0.1 cm, and for a gas pressure of 10^{-2} torr, the error in calculating $(1 - A)$ is 6.5%. The error is proportional to $1/n$, so that for 500 slabs the error is only 1.3%. The error is due to the assumption regarding the numbers of atoms lying behind one another in the same slab.

Since we are interested in those particles with final velocities $> 1 \times 10^5$ cm/sec, regardless of what happens on the way to the substrate, it is possible for some particles to make more than one collision and still have a velocity $> 1 \times 10^5$ cm/sec. Some may make as many as 10 collisions before their velocity falls below the lower limit. It is therefore necessary to consider collisions in each slab, whence the larger fraction of atoms will continue with the same velocity and a small fraction will set out with a velocity modified by the collision in the slab. If the fraction of atoms which have had a collision is plotted as a function of n , a set of curves is obtained whose shape 372

and starting points depend upon the velocity and the position of the slab in which collision occurred.

Since the collision cross-section is a function of velocity, this must also be taken into account. From thermal beam scattering, q varies as $v^{-2/5}$, assuming an r^{-6} variation of potential with distance [13]. If we assume this law, and if a value for the collision cross-section at 0° C is taken as $\frac{1}{2}(D_1 + D_2)$, where D_1 and D_2 are the molecular diameters of silver and argon respectively, the value of q as a function of velocity may be calculated.

The average energy loss per collision by a molecule, f , is given by Cravath [14] for the case of two gases with Maxwellian distributions corresponding to temperatures T_1 and T_2 respectively.

$$
f = \frac{8}{3} \frac{m_1 m_2}{(m_1 + m_2)^2} \left\{ 1 - \frac{T_1}{T_2} \right\}
$$

where m_1 , m_2 are the masses of the colliding particles.

For the case of silver in argon, $T_1/T_2 \ll 1$ and $f \approx 0.5$; hence the average fractional velocity loss by a silver particle is $(1 - 1/\sqrt{2})$.

A computer programme has been written to calculate the fraction of atoms arriving at the substrate with velocities $> 1 \times 10^5$ cm/sec, using the curve given by Wehner as the starting distribution. The results of the computation are shown in fig. 3 for various gas pressures and a target-substrate separation of 10 cm, divided into 500 slabs. A value of 33.5×10^{-16} cm² was taken as the collision cross-section, assumed to be measured at 0° C, i.e. a relative velocity of 5×10^4 cm/sec. This is equal to $\frac{1}{2}(D_1 + D_2)$ as indicated above.

The calculations show that, at a pressure of 1×10^{-2} torr, a maximum of 15% of all particles which start with velocities $> 1 \times 10^5$ cm/sec (0.6 eV) arrive at the substrate with velocities greater than this value. Fig. 4 shows how this percentage changes with pressure. It must be emphasised that these figures are maximum values, since it has been assumed that all particles continue in a forward direction following collision.

These results indicate that there will be a significant difference between the arrival energies of sputtered and evaporated atoms even for sputtering pressures of 10^{-2} torr. This difference may influence the growth behaviour in two ways. The arriving atoms may cause secondary sput-

Figure 3 Calculated **velocity distribution of silver atoms arriving at the substrate for various argon gas pressures** (10 cm cathode-substrate spacing).

Figure 4 Calculated variation of percentage of **silver atoms** arriving **at substrate with energies above** 0.6 eV as a function of argon **gas pressure** (10 cm **cathode-substrate spacing).**

tering of impurities on the surface of the substrate, or they may penetrate the substrate and create nucleation sites that are intimately associated with the substrate.

4.3. Effect of Cleanliness of Substrate

Exposure to a glow discharge is commonly used as a method of cleaning substrate surfaces. Maissel and Schaible [15] employed a technique of bias sputtering in which the substrate was biased \sim 100 V negative, and found that the purity of tantalum films deposited on glass was higher than that for films deposited by non-bias means. The reason for this was that positive ions were attracted to the substrate continuously during deposition, and impurity atoms were sputtered preferentially by these ions. In fact, they found the cleaning of the substrate to be so effective that films would not adhere. Deliberate oxidation of the first few layers of the film was employed so as to make the films stick.

The voltages employed by Maissel and Schaible would cause the positive singly-charged ions striking the substrate to have a maximum energy of some 100 eV, but, because of collision processes at the gas pressures used, many will have energies much less than this.

The cleanliness aspect has been investigated in the case of evaporated films of gold on rocksalt. Matthews and Grunbaum [16] find that, on vacuum cleaving a rocksalt substrate at 10^{-9} torr, and subsequent deposition of gold, several orientations occur, the least likely of which is the (I00). This orientation requires exposure of the substrate surface to moist oxygen or moist air for it to grow preferentially [17]. Jaunet and Sella [8] have found that, for silver on rocksalt, the critical temperature for epitaxy is reduced from 150 to 20° C for the (100) orientation, when the substrate is cleaved under vacuum at $10⁻⁶$ torr. With a range of evaporants, they report a lowering in epitaxial temperature of about 150° C if the substrate is vacuum cleaved.

From the results of these authors, it would appear that a clean surface is desirable for the growth of single crystal (100) films on (100) rocksalt at low temperatures, though a certain amount of particular contaminants does appear to be necessary. It may be, therefore, that the sputtering action does partially clean the substrate, but the water and oxygen contaminants are not removed as effectively as other impurities. Hence the preferred growth of (100) material.

4.4. Surface Penetration

The penetration of a substrate by high-energy particles is well known [18]. At the highest arrival energies, a molten zone is created at the arrival point [19], while at lower energies the particles will simply penetrate the substrate. It is to be expected that some penetration will occur at least down to the sputtering threshold. This threshold may be as low as 20 eV [20]. Still lower arrival energies could give small penetration of the lattice without actual sputtering. It has been shown [21] that arrival energies of 1 eV compared with 0.02 eV do not affect the growth process of gold on rocksalt, but it is possible that the arrival energies of a percentage of the atoms in the experiments reported here may be sufficient to penetrate the surface of the rocksalt, as some will lie in the range 1 to 20 eV.

4.5. Charged Particle Effects

The second difference that was noted between evaporated and sputtered deposition was the existence of negative ion or electron bombardment of the substrate. In our experiments, the films were prepared on the anode of a diode system, so that the substrate was being bombarded continuously by electrons at energies up to 3 keV. The number of negative ions will be very small. However, electron currents would be of the same order as the ion current; i.e. in terms of numbers, $\sim 1 \times 10^{14}$ electrons/cm² sec. Stirland [22] has shown recently the effect of electrons on the degree of orientation of the film. In a gold evaporation experiment, a rocksalt substrate was maintained at 150° C and was vacuum cleaved so as to expose two similar cleavage faces. One crystal face was protected while the other was bombarded with 75 eV electrons at a density of 1×10^{14} electrons/ $cm²$ sec. It was found that the orientation of films of any thickness was markedly different. The non-bombarded crystal was polycrystalline with (100) and (111) preferred orientations, while the bombarded crystal was perfectly oriented (100).

The numbers of electrons involved in these experiments are similar to those encountered in our sputtering experiments. The major difference is the energy of the bombarding electrons. The substrates used to-date have been ionic crystals, and further examination of deposition onto other substrates under electron bombardment conditions is necessary. As far as sputtering is concerned, we are currently examining low-pressure sputtering in which an independent source of 374

electrons is used, and the ions are extracted from the discharge at right-angles. The number of electrons striking the substrate is thus much reduced, and may be completely eliminated by use of an electron reflector.

5. Discussion

5.1. Rate and Temperature Effects

It has already been mentioned that, in the sputtering arrangement used in the present work, rate variation was achieved by varying the gas pressure. The consequent change in the number of higher energy particles arriving at the substrate (energies > 0.6 eV, i.e. $v > 1 \times 10^5$ cm/sec) as a function of rate of deposition is shown in fig. 5, and it can be seen that the numbers fall off rapidly with increase in rate above 0.5 Å/sec. Thus, if epitaxy is associated with the arrival of high-energy particles, as suggested in sections 4.3 and 4.4 above, then one would predict that at high rates there would be very few oriented nuclei, and a polycrystalline film would result. However, fig. 5 suggests that the dependence of orientation on incident rate would fall off sharply.

Figure 5 Calculated variation of percentage of silver atoms arriving at substrate with energies above 0.6 eV as a function of rate of deposition (10 cm cathode-substrate spacing).

The observed requirement for higher temperatures in order to obtain epitaxy at higher rates may be due to the desorption of contamination. Since the energy of arrival of the sputtered material is reduced at high rates, the activation energy for desorption of the contaminant would have to be supplied by the substrate temperature. Increasing rates would therefore require increasing substrate temperatures.

Since there is little available data [23] for the deposition rate versus substrate temperature relationship for silver evaporated onto rocksalt, we are unable to say whether the curve obtained in fig. l only shows a displacement from the higher temperatures required for epitaxy by evaporation, or also shows a change in slope. A displacement of the curve can be discussed in the terms given above, but a change in slope would imply a change in the energetics of the nucleation and growth process.

5.2, Thickness Effects

In discussing the dependence of orientation on thickness, one is not dealing with a situation peculiar to sputtering, whereas both surface penetration and charged particle effects are features of sputtering alone.

Matthews and Allinson [24] have observed an orientation dependence on thickness for gold evaporated onto rocksalt. They show that large crystallites are better aligned than small ones, and they argue that, when two crystallites coalesce, the smaller of the two will rotate to be in parallel or twin relationship to the larger. There will thus be an increase in alignment of the crystallites in the film. However, they observed that this alignment does not necessarily lead to single crystal films. Matthews *[25]* has recently shown that it is possible to consider the preferential growth of certain orientations. Thus, under certain conditions, the initial crystallites in these orientations will grow more rapidly than others, and eventually the whole film will be of one type. The actual crystal planes that are preferred and their rates of growth will depend upon the energetics of the system, and the presence of impurities can play a considerable part. Pashley [26] has also observed orientation dependence on thickness for chemically grown films.

It appears therefore that some orientations grow at faster rates than others, and thus a thickness-dependent orientation is obtained. If it is postulated that the nuclei at sites which are intimately connected with the lattice – either due to the substrate being clean or to surface penetration - grow at faster rates, then an oriented film will be obtained dependent upon the number of initially oriented nuclei.

5.3. Effect of Electrons

The evidence of the effect of electrons on deposition by evaporation has been experimental, and no explanation has yet emerged. It might be expected in sputtering that higher rates of growth, which imply higher electron densities, should show enhanced epitaxy. This is the reverse of the observed effect. If the energy of the electrons is an important factor, then the higher gas pressure associated with increased rates would result in a slowing down of electrons due to increased collisions, and the films would be less perfect. However, this process would not be expected to be as sensitive to change in rate as that involving the neutral, sputtered atoms since, following collisions, an electron is accelerated by the applied field.

6. Conclusions

(a) It has been established that it is possible to grow single crystal silver films on rocksalt, by sputtering in a glow discharge, at appreciably lower temperatures than by evaporation.

(b) A dependence of perfection of orientation on rate of deposition, substrate temperature, and film thickness has been observed. Within the range considered, epitaxy is observed for the lower rate of deposition, the higher substrate temperature, and the thicker films.

(c) The results have been discussed in terms of the energy of arrival of the sputtered particles at the substrate. At the pressures employed in the experiments (approximately 1×10^{-2} torr), it has been shown that the numbers arriving with significantly high energy are between 1 and 30 $\frac{\%}{\%}$, and it is possible that the observed rate and temperature dependence of epitaxy is due to surface cleaning and penetration effects.

(d) Thickness effects have also been discussed in terms of the arrival energy, and it is postulated that at the lower rates and higher substrate temperatures more oriented nuclei are produced than when high rates and low substrate temperatures are employed. These oriented nuclei then grow at faster rates than the unoriented ones do.

(e) The effect of arrival of negative ions or electrons on the surface has been considered. Experiments by other workers using evaporated gold have shown the significance of electron bombardment which may be present in the sputtered case.

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