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\(\text{Å/sec}\), films have been grown with \((100)_{\text{Ag}} // (100)_{\text{KCl}}\) orientation, at temperatures in the range -35 to 0° C. Higher rates required higher temperatures. Films giving these results were all 200 Å in thickness. A thickness dependence of orientation has been observed for films below 120 Å 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.

1. 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 epitaxial 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 12 in. diameter × 14 in. high, glass chamber evacuated by a 4 in., oil diffusion pump backed by an oil rotary pump (1 in. = 2.5 cm). With liquid-nitrogen 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 1.10^-2 torr, but in all cases the system was evacuated to <2.10^-6 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 NaCl in air, to reveal a (100) 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 μA/cm² 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 Å/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 flat 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, stainless-steel 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 ~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 Å.

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