THE EPITAXY OF GOLD

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Previous studies of the epitaxial growth of gold by vapor deposition on a variety of substrates have been compiled and listed according to the type of substrate. A short introduction describing the epitaxy process has been included for general reference purposes and some highly promising applications of thin film technology are discussed.

Introduction

Epitaxy, in the usual sense of the word, is a term that refers to the oriented growth of one material, the overgrowth, on a crystalline substrate. The orientation relationships are such that the {HKL} plane of the overgrowth is parallel to the {hkl} plane of the monocrystalline substrate. Correspondingly the <UVW> and <uvw> directions are parallel. The relationships are usually written as follows: {HKL} // {hkl} and <UVW> // <uvw>. Exactly what those relationships are can often be predicted on the basis of lattice fitting or lattice misfit type arguments. Thus, those planes and directions which give the best lattice fit generally determine the orientation of the film with respect to the substrate. The misfit that occurs produces strain, which, if large enough, may generate line defects called misfit dislocations at the interface between the film and the substrate. These dislocations tend to reduce the misfit strain. The theoretical basis for this result will be outlined in a subsequent section.

Investigations of epitaxial growth phenomena are an important segment of thin film studies. The processes that are involved in forming an epitaxial overgrowth may involve the solid, liquid, and vapor states. Thus, the growth of solid phases in alloys may involve interfacial epitaxial relationships. In the liquid state one can form epitaxial overgrowths by electrodeposition or by a process called liquid phase epitaxy (LPE), whereby a saturated solution plates out a material on a particular solid substrate.

Vapor phase methods are probably the most popular and include:

- Vacuum evaporation from a hot source onto a colder substrate (nowadays more popularly called molecular beam epitaxy - MBE),
- Chemical vapor deposition (CVD), which involves surface chemical reactions of gases at pressures usually not too different from atmospheric (e.g. thermal decomposition of gas on a hot substrate or polymerization of monomers), and
- Ion sputtering processes whereby ions produced in a gas discharge (or by other gaseous ionization methods) are accelerated towards a target. The subsequent interaction by momentum interchange results in the emission of atoms or molecules from the target material which then are permitted to strike a substrate on which the film of interest grows.

Several other methods also have been employed to form thin films but these are not as useful in epitaxy studies as the ones described above.

The properties of epitaxial films can be made to vary widely because of the high reactivity of individual atoms and molecules. Thus in combining the atoms to form a thin film, numerous physical and chemical processes may be involved, thereby making possible an almost limitless variety of properties, for example, microstructure (i.e. defect content), orientation, composition, and topography. This result is especially true for very thin films. In this case their

physical, chemical, and mechanical properties may be widely different from those of either the bulk material or the substrate.

Recent studies on surface catalytic reactions, for example, show that the catalytic reactivity of thin epitaxial films, approximately one monoatomic layer thick, formed on particular metal substrates may significantly increase the reactivity of the surface for a particular chemical reaction. Typical examples are Au on Pt (1), and Cu on Ru (2). Since bulk gold and bulk copper are normally quite unreactive in the cases in question, this result is very surprising. Clearly the reverse effect, namely the reduction of surface reactivity by a more reactive thin film material, should also be possible with enormous implications in the field of corrosion. Thus by using thin film techniques, one can devise an almost limitless range of new material surfaces, each of which having its own distinct surface chemistry.

In the case of electrical properties, it has been shown that an infinite variety of band structures can be prepared by making periodic, thin layers of films and stacking them on top of each other. These so-called 'strained-layer-superlattices' (formed by MBE) provide the semiconductor industry with the opportunity to tailor-make particular microelectronic devices that simply would not be possible using only bulk materials (3). As a consequence the range of possible microelectronic devices has been enormously extended.

Epitaxial growth of thin films will allow the current more or less planar integration, common in microelectronic devices, to be extended into three dimensions, thereby reducing the space needed for the associated devices. To accomplish this task one needs to be able to grow epitaxial metal and especially insulator films on silicon (or, in general, on other semiconducting substrates, such as GaAs) as well as epitaxial silicon layers on epitaxial insulator substrates. Great efforts are currently being expended to form these epitaxial Si/insulator/Si/insulator/Si... multilayers with the degree of perfection that is required.

These few illustrations show that epitaxy studies represent not only interesting science but also have enormously important applications in industry, especially in the areas of microelectronics, catalysis and corrosion.

The present paper reviews the work that has been reported on the epitaxial growth of gold films formed on a

Fig. 1 Schematic illustration of the basic thin film growth modes: (a) layer (FVM), (b) layer by layer, (c) layer plus island (SK), (d) island (VW).

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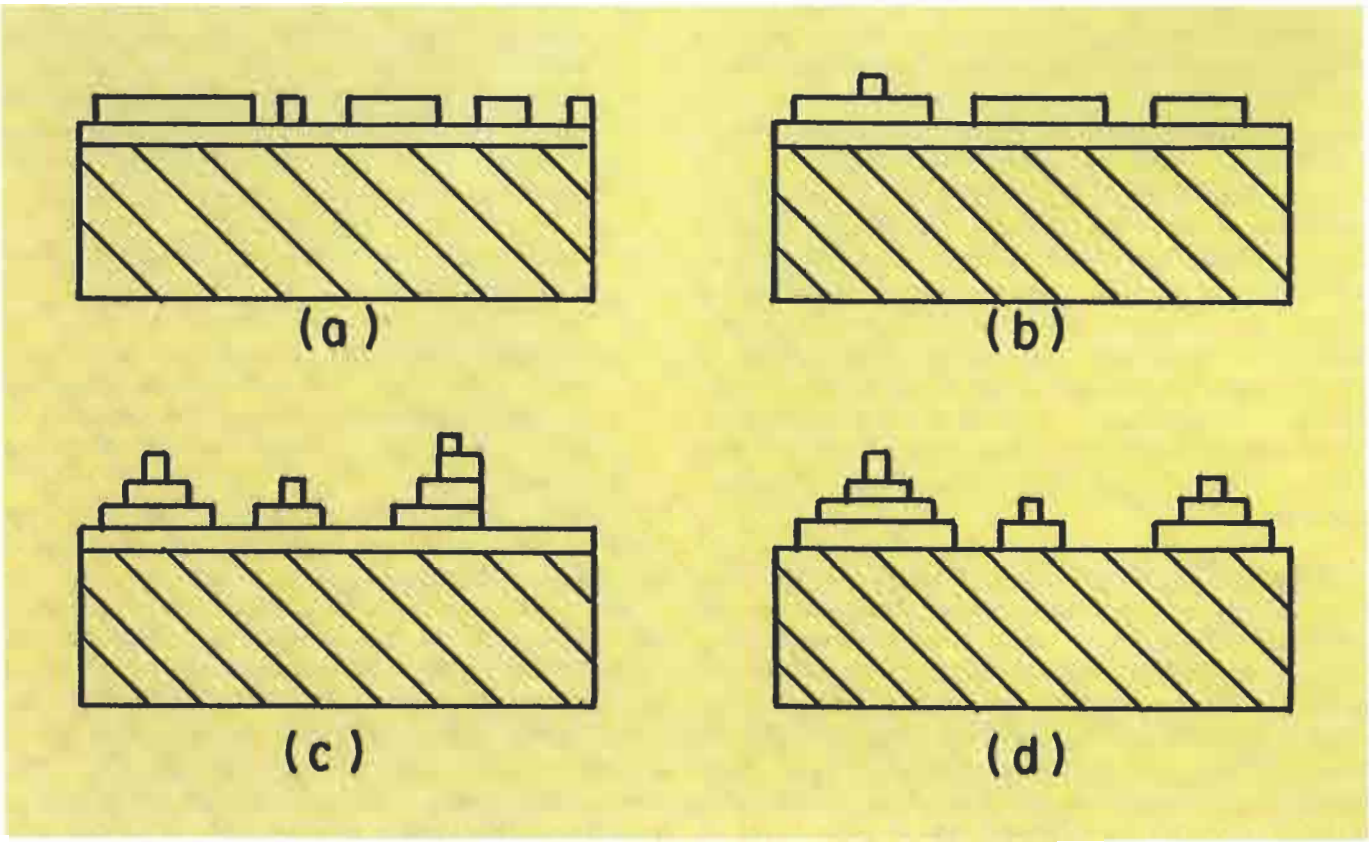

variety of substrates. A listing of sources is presented in a series of tables that are up-to-date through early 1986. This work builds on two previous reviews published in the early 1970s (4, 5). The current compilation should serve as a useful reference for previous studies on the epitaxy of gold films.

Epitaxial Film Growth Modes

The formation of a thin film by any of the experimental methods mentioned earlier is not a simple process. Generally atoms or molecules which are incident on a surface may either diffuse over the surface, diffuse into the bulk of the substrate, or be desorbed. In the latter case no film forms. In typical cases one or more of these processes dominates the film formation process. Au films, for example, deposited on most substrates at temperatures at least within 100K and 1000K, surface or bulk diffuse before they become bound to their final resting place. Desorption is negligible and films form easily.

The growth mode of a film, however, falls into one of three basic categories:

- Layer (Frank and van der Merwe (6): FVM),
- Layer plus island (Stranski-Krastanov (7): SK), and
- Island (Volmer-Weber (8): VW).

These growth modes are illustrated in Figure 1, which also shows a modification of the layer mode called 'layer-by-layer' (Figure 1b). In this latter case the $(n+1)$th layer starts growing before the nth layer is complete. Thus normal layer growth means that monoatomic layers form on top of each other, one at a time. In this case, incident atoms surface diffuse until they find a step where they attach themselves, thereby extending the step. Clearly, surface imperfections and impurities may have a drastic effect on this crucial surface diffusion process.

In the SK growth mode, one or several layers form first. Then island nuclei form and grow on top of the initial layers. In the VW mode, island nuclei form initially on the substrate, increasing their density and sizes as deposition continues until they coalesce. Further deposition results in a completely continuous film that may eventually become quite flat. On this flat, thick film surface, further film growth by a layer mode may occur.

Thermodynamic considerations (for a recent review see Reference 9), modified by supersaturation considerations have been used to predict the growth mode in particular cases. The theory is based largely on surface and interfacial energy considerations. The surface or interfacial energy E_{hkl} can be defined in terms of the work W_{hkl} needed to separate a homogeneous crystal along a particular crystallographic plane ($E_{hkl} = \frac{1}{2}W_{hkl}$) or along the interface between two different crystals. Generally, materials with low surface energies, when deposited on high surface energy substrates, will grow in a layer growth mode because

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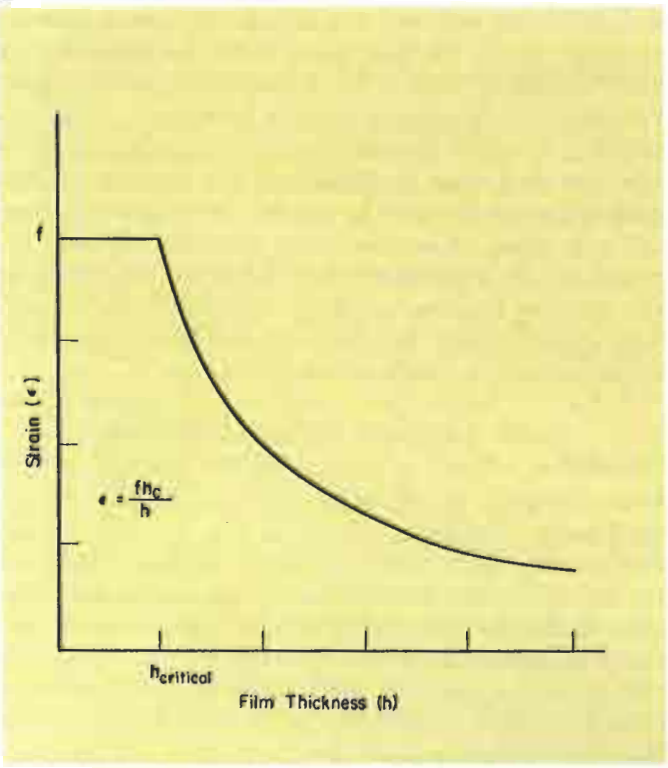

Fig. 2 The relationship between strain in an epitaxial cubic film and its thickness.

the total energy of the system will be reduced. In the reverse case, where a high surface energy material is grown on a low surface energy substrate, island growth is favoured. SK growth is predicted if the conditions for layer growth are complemented by a high lattice strain energy at the interface caused by a large misfit in the lattice parameters, and/or the presence of quite different structures in the two crystals.

Interfacial Strain and Misfit Dislocations

The strain in a film growing by a layer growth mechanism has been derived (10). Figure 2 illustrates the results. Up to a critical film thickness, h_c, the strain equals the misfit, which for a simple cubic crystal is given by

$$f = (a_s - a_f)/a_f$$

where a_s and a_f are the unstrained lattice parameters of the substrate and film respectively. Thus positive (negative) f values mean that the film in the interfacial region experiences a tensile (compressive) strain. Generally one assumes a rigid substrate, especially for very thin films. The formula for the curve in Figure 2 for $h > h_c$ is complicated but shows that the strain decreases as h^{-1}. In this same region, misfit

dislocations are introduced at the interface and they are responsible for the reduced strain. Their function is to localise the misfit strain to the regions of the misfit dislocations, leaving wide areas of good fit in between. As the film thickens, the misfit dislocation density increases, rapidly at first, and then more gradually until the strain in the film goes effectively to zero. In practice, measurements show that this strain elimination takes place in less than approximately 10 monolayers. Such effects are also present at the interface between an island overgrowth and its substrate, but of course the effects are modified by the size of the island and the interfacial area of contact.

Gold-Substrate Epitaxial Systems

This section will list the various epitaxial systems that have been reported in the literature. Extensive use of two previous listings were made, the earlier one by Gebhardt and Neuhaus (4), which covered systems from 1836 to 1970, and the list by Grunbaum (5) covering systems through July, 1974. In the listing by Gebhardt and Neuhaus the orientation relationships are given, while that by Grunbaum indicates the experimental methods used to study the particular epitaxial systems. In the present list, only the overgrowth (Au) and substrate material are given along with the references. The first element (A) in the A/B notation is the overgrowth (Au) and the second, B, is the substrate. The various systems are listed according to the types of substrates used.

Acknowledgement

This work was supported in part by a grant from the United States Department of Energy, Basic Energy Sciences Division, on grant number DE-FG02-84ER45139.

Table I
Au/Metals

Substrate	References
Ag	11-52
Al	53
Cr	54, 55
Cu	12, 56-71
Cu_3Au	72
Fe	56, 57, 73
Mo	74-79
Ni	19, 80
Pb	81, 82
Pd	12, 19, 27, 45, 50, 63, 82-92
Pt	12, 93-96
W	65, 78, 97-104
Zn	105

Table II
Au/Selected Semiconductors and Chalcogenides

Substrate	References
C (graphite)	106-110
Ge	111, 112
Si	65, 111-125
GaAs	126-130
GaSb	131-133
MoS_2	45, 82, 109, 110, 134-149
PbS	65, 150-154
PbSe	65, 153, 155
PbTe	65, 153
SnTe	153
ZnS	80

Table III
Au/Carbonates, Oxides, Mica

Substrate	References
$CaCO_3$	12, 57, 156
Al_2O_3 (sapphire)	157, 158
$BaTiO_3$	159, 160
MgO	45, 65, 109, 110, 145, 160-174
SiO_2 (quartz)	11, 57, 156, 160
ZnO	175, 176
Mica	12, 34, 37, 57, 148, 156, 160, 172, 177-191

Table IV
Au/Metal Halides

Substrate	References
CaF_2	57, 156
CdI_2	192
KBr_2	107, 193-201
KCl	65, 107, 166, 193, 196-200, 202-227
KF	198, 228
KI	194, 196-199, 202-204, 206, 209
LiF	65, 168, 178, 195, 198, 202, 229
NaBr	198, 200
NaCl	11, 12, 14, 16, 33, 40, 57, 65, 87, 93, 107, 159, 162, 173, 182-184, 187, 188, 193-196, 198-200, 202-206, 209, 212, 214-217, 224-226, 229-320
NaF	198, 200
NaI	198
$PbCl_2$	276
$RbBr_2$	198
RbCl	198
RbI	198

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