Review of the fundamentals of thin-film growth

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The properties of a thin film of a given material depend on the film's real structure. The real structure is defined as the link between a thin film's deposition parameters and its properties. To facilitate engineering the properties of a thin film by manipulating its real structure, thin-film formation is reviewed as a process starting with nucleation followed by coalescence and subsequent thickness growth, all stages of which can be influenced by deposition parameters. The focus in this review is on dielectric and metallic films and their optical properties. In contrast to optoelectronics all these film growth possibilities for the engineering of novel optical films with extraordinary properties are just beginning to be used. © 2002 Optical Society of America

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1. Introduction

The dependence of optical properties on deposition conditions is a severe constraint in reproducing thin film's performance. There is a significant conflict between performance models based on ideal- and real-structure films. Models are required that relate real-film structure to desired optical properties. This knowledge would permit the design and manufacture of coatings based on more-realistic production conditions than are now possible. In this review aspects of thin-film growth in various dimensions, from isolated nanoclusters to continuous macroscopic films, are treated. The focus is on dielectric and metallic films and their optical properties.

Investigations of process parameters, real structures, and film properties have been in progress for many years, starting in 1938 with the classic research of Hass.[1] Perusal of Refs. 2–7 is recommended as an introduction to the structure-related optical properties of thin films.

2. Real Structure

The ideal structure of solids is either ideally ordered single-crystalline or ideally disordered amorphous. But there is no ideal crystalline or amorphous structure in the real world. The real structure of single crystals always has a certain number of intrinsic and extrinsic defects. Intrinsic defects are those that are caused by atoms' not changing the stoichiometry of the crystal. Defects that produce nonstoichiometry are called extrinsic defects. Defects generally can have a major influence on the physical properties of the solid, for instance, on the transmission of light and on electrical conductivity.

The growth of thin films by physical-vapor deposition is an extremely nonequilibrium process that takes place at high supersaturations and at comparatively high concentrations of impurity atoms. Consequently the number of extrinsic and intrinsic defects in thin films may exceed that in bulk solids by ~5 orders of magnitude. As a result of the usual deposition conditions in optics and of the surface structure of optical components, optical films are generally polycrystalline. This means that the real film consists of defect-rich crystalline grains, which are separated by impurity-enriched grain boundaries. This is the reason why film properties differ significantly from bulk properties. Figure 1 shows this fact schematically. The real structure of thin films is a metastable nonequilibrium state that is far away from its energetic minimum. Real structure tends to change whenever enough energy is available.

Real structure is the link between thin-film deposition parameters and thin-film properties (Fig. 2). In engineering electronic and optical properties it is vital to control stoichiometry, mean grain size, and grain size distribution of the thin-film structure. For engineering of film properties through real structure, models are indispensable. In what follows, thin-film formation is described as a kinetic adsorption and diffusion process starting with nucleation, coalescence, and subsequent thickness growth,

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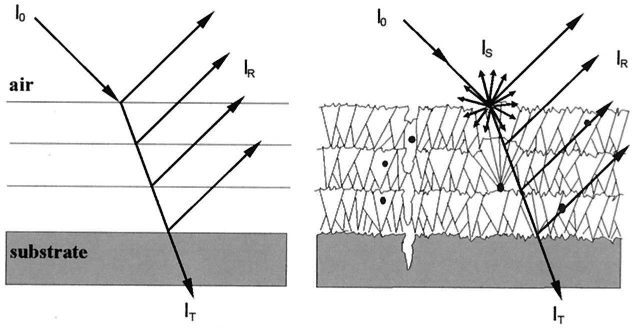


Fig. 1. Left, ideal single-crystalline substrate coated with ideal single-crystalline films. Incoming light with intensity I_0 is split into reflected and transmitted parts I_R and I_T, respectively. Conservation of energy is given by $I_0 = I_R + I_T$. Right, real substrate with real coatings. Part of incoming intensity I_0 is absorbed (I_A) or scattered (I_S). Conservation of energy is given by $I_0 = I_R + I_T + I_A + I_S$.

whereby all states can be influenced by deposition parameters.

3. Growth of Thin Films

A. Nucleation

The production of thin films by vapor deposition is a classic case of heterogeneous nucleation, i.e., the condensation of adatoms at a substrate that is composed of atoms that are different from those of the vapor. Figure 3 shows the various stages of thin-film growth.

deposition process (temperature, rate, energy, residual gas)
substrate surface structure → ↓
real structure (grain size, orientation, defect density)
↓
properties (complex refractive index, stress, hardness, …)
↓
application (antireflection, high reflection, filtering)

Fig. 2. Factors that control the properties of thin films.

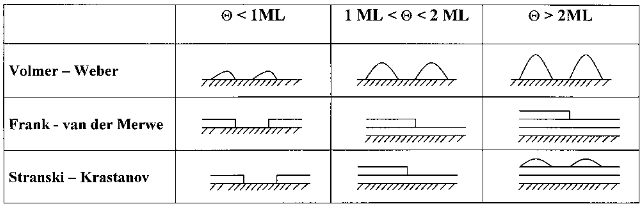


Fig. 4. Initial states of film growth (after Ref. 9). Θ, substrate surface coverage in monolayers (ML).

Nucleation takes place at high supersaturations S, defined as

$$S = p/p_e, \tag{1}$$

where p is the vapor pressure of the deposit material evaporated from the source at temperature T and p_o is the equilibrium vapor pressure of the substrate material at temperature T_S. Deposition rate R is related to p as

$$R = p/(2\pi mKT)^{1/2}, \tag{2}$$

K is Boltzmann's constant, m is the molecular weight of the deposit material, and T is the temperature of the evaporation source. Because S is extremely high, $S = 10^5$–10^{50} ($S \approx 4$ in a Wilson chamber), nucleation is a nonequilibrium process, which can best be described by the mean-field nucleation theory.[8]

As for all phase transitions, the formation of thin films is characterized by the formation of nuclei and their growth. Depending on the interaction energies of substrate atoms and film atoms, any of three growth modes (Fig. 4) can occur:

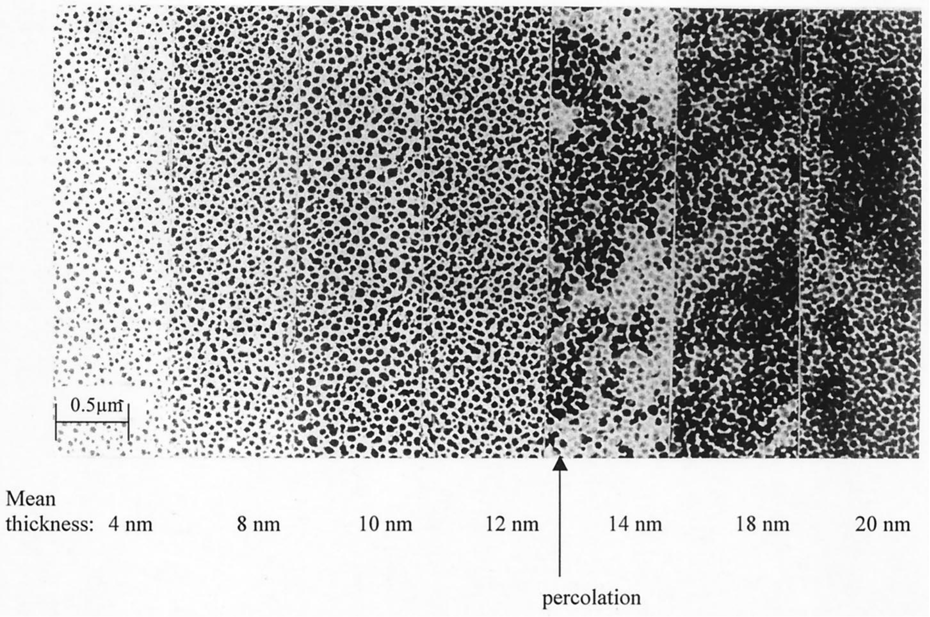


Fig. 3. TEM micrographs of antimony film formation (nucleation, growth of nuclei, coalescence, channels, holes, homogeneous film). Here (as is special for Sb) at percolation an amorphous–crystalline phase transition (change of contrast) takes place.

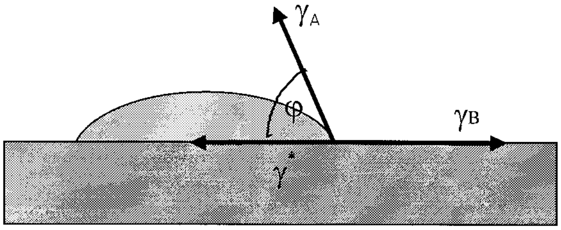


Fig. 5. Wetting angle φ of a liquid nucleus on a substrate is described by Young's equation: $\gamma_B = \gamma^* + \gamma_A \cos \varphi$, where γ_B is the surface energy of substrate, γ_A is the surface energy of film material, and γ^* is the interface energy film–substrate.

- *Layer by Layer* In the two-dimensional Frank–van der Merwe mode, layers of material grow one on top of another. Interaction between substrate and film atoms is greater than between adjacent film atoms.
- *Island* In the Volmer–Weber mode, separate three-dimensional islands form on the substrate. Interaction between film atoms is greater than between adjacent film and substrate atoms.
- *Layer plus Island* In the Stranski–Krastanov mode, one or two monolayers form first, followed by individual islands.

Growth modes can be systematically classified in terms of surface energies with Young's equation taken into account (Fig. 5).

Island growth ($\varphi > 0$) requires that $\gamma_B < \gamma_A + \gamma^*$, whereas layer growth ($\varphi = 0$) requires that $\gamma_B > \gamma_A + \gamma^*$. Layer-plus-island growth occurs because the interface energy increases with film thickness; typically the layer on top of the substrate is strained to fit the substrate. The growth mode is controlled not only by interface energies but also by supersaturation.[10] Generally, growth tends to shift from island to layer as supersaturation increases.

B. Coalescence

The next stage of three-dimensional film formation is the growth of islands until they touch one another to form a continuous network. This so-called coalescence process is extremely important for the design of films with special properties. After solidlike coalescence of two islands there may remain a grain boundary between them, or they may fuse together in a liquidlike fashion to form a new, larger, and boundary-free island. Surface energies and supersaturation are the factors that control these processes through material transport by surface and bulk diffusion. The transition from isolated islands to a continuous macroscopic network can be characterized by a percolation threshold thickness. To percolate means to pass or to cause to pass through a porous material. This definition can be understood, for example, from measurements of electrical conductivity between two macroscopically separated contacts upon an insulating substrate during the growth of a metallic layer between them. At percolation thickness, coalescence creates a continuous path for electrons through the network and conductivity rises steeply by several orders of magnitude. Obviously, a phase transition takes place at the percolation threshold, with optical film properties changing exponentially. It is shown that this phenomenon is crucial when it comes to engineering thin semitransparent metal films.

C. Thickness Growth

The mean percolation thickness for many films is 1–20 nm. The thickness required for producing metal mirror layers or dielectric quarter-wave systems is, however, larger than that value. Further material deposition does not simply increase thickness; moreover, the real structure is determined by the following four processes:

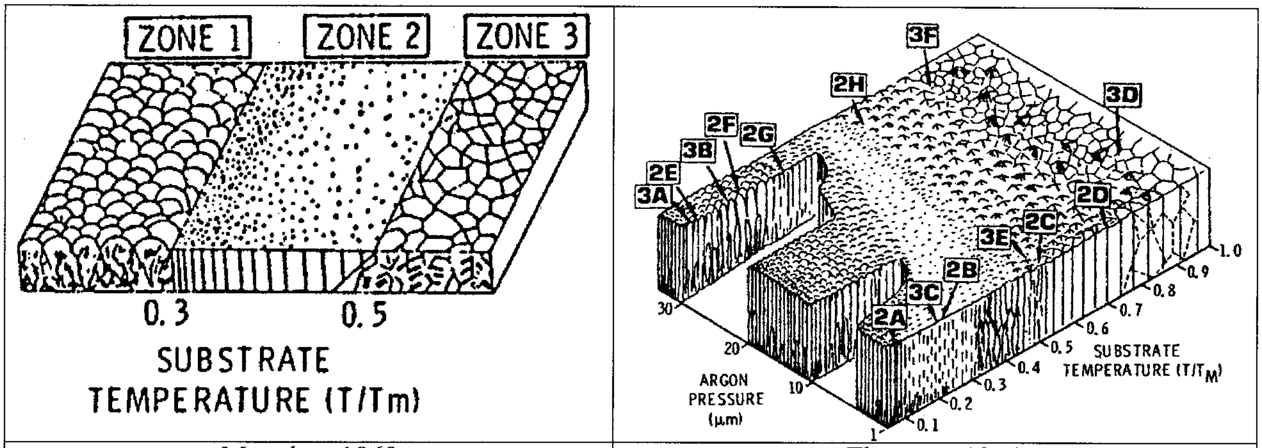
- *Shadowing* A geometric interaction between the arriving admolecules and the roughness of the growing surface. This effect is dominant at low substrate temperatures T_S. It occurs because the vapor beam is directed.
- *Surface diffusion* Mobility of admolecules at surfaces and interfaces such as grain boundaries; dominant at medium substrate temperatures T_S.
- *Bulk diffusion* Mobility of admolecules in the volume of grains; dominant at high T_S.
- *Recrystallization* Phase transition as a complete change of crystal orientation; dominant at percolation thickness, large film thickness, and high T_S.

For most metals and dielectrics, activation energies for diffusion are related to the melting temperature of the material, T_m. Thus different basic processes can be expected to dominate different ranges of T_s/T_m and to determine the real structure. This is the concept of so-called structure zone models.[11] In the simplest case, real structure exists in three zones:

- Zone I $T_s/T_m < 0.3$ (low mobility; admolecules stick where they land; the result is a fine-grained porous real structure).
- Zone II $0.3 < T_s/T_m < 0.5$ (surface diffusion occurs with activation energies of 0.1–0.3 eV; a columnar real structure is obtained).
- Zone III $T_s/T_m > 0.5$ (bulk diffusion occurs with activation energies above 0.3 eV, resulting in a rough equiaxed grained real structure).

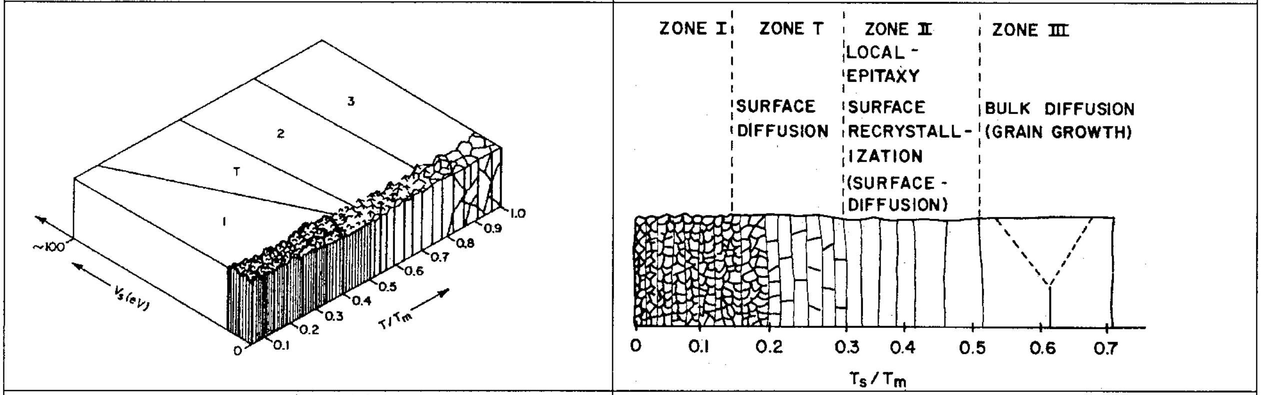
Such a generalization of homologous structure zone models in terms of physical processes is important for engineering film properties. Structure zone models have been deduced by Thornton for sputtered films[12–15] and by Messier *et al.*[16] and Messier[17] for ion-assisted deposition. Additionally, Grovenor *et al.*[18] take the influence of substrate morphology into consideration. For comparison, all models mentioned are shown in Fig. 6.

Until now it has only briefly been mentioned that the growth of thin films for optics by physical-vapor deposition is a comparatively dirty process. Indeed, under high-vacuum conditions in a normal produc-



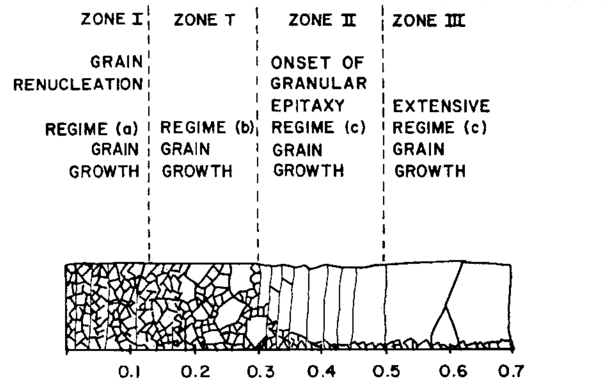
Movchan 1969

Thornton 1974



Messier 1984

Grovenor 1984



Grovenor 1984

Fig. 6. Structure zone models (after Movchan,[11] Thornton,[12] Messier *et al.*,[16] and Grovenor *et al.*[18]).

tion environment the rate of residual gas molecules (water, hydrocarbons, etc.) striking the substrate surface is of the same order of magnitude as the deposition rate of the film material. At a total pressure of 10^{-4} Pa it takes only $\sim$1 s to form one monolayer of residual gas molecules upon the substrate. Consequently, extrinsic impurity defects, whether present on the substrate surface or resulting from the evaporation source or from the residual gas, have a great influence on film growth. The effect is comparable with that of the substrate temperature. Impurities markedly change the surface energies at all stages of film growth. Impurities that are not soluble in the lattice are segregated into the grain boundaries and onto the growing film surface. At a critical impurity concentration, a fixed passivation layer forms, upon which secondary nucleation may occur. Therefore, nondiluted impurities are concentrated at interfaces.

In real structures, large impurity concentrations act in the same way as low substrate temperatures. Structure zones are shifted along the T_s/T_m axis toward a fine crystalline, porous structure. Barnal and Adamik[19] introduced the basic structure zone model, which starts from the basic case of impurity-free films (Fig. 7) and then takes the increasing influence of impurities into consideration.

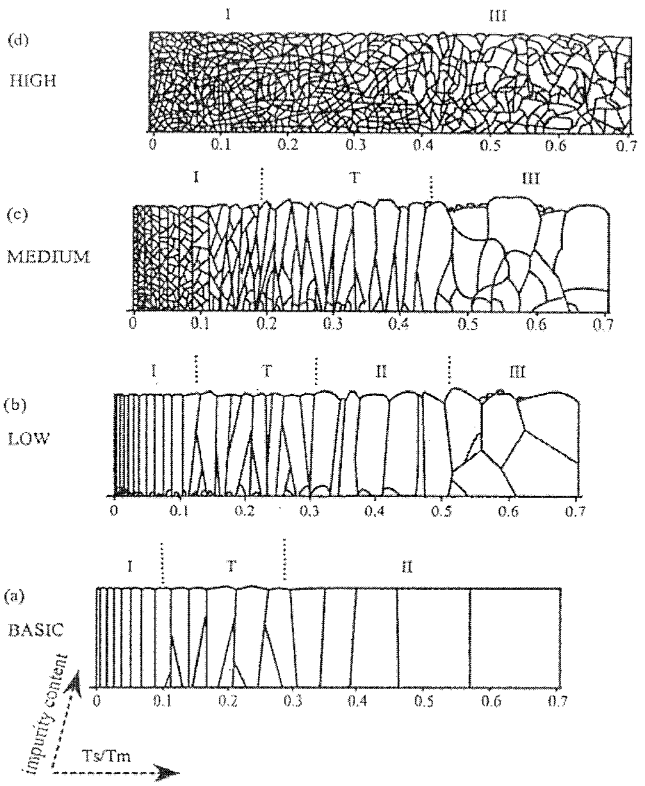


Fig. 7. Basic and real-structure zone models for low, medium, and high impurity concentrations (after Barna[19]).

4. Dielectric Films

Classic optical interference film systems are now applied down to the vacuum-ultraviolet excimer-laser wavelength range. Here the individual quarter-wave fluoride films can be as thin as ~10 nm. Structure zone models (Figs. 6 and 7) show the real structure of dielectric thin films. The connection between optical properties and real structure is possible by effective medium theories on the basis of the classic Lorenz–Lorentz model, e.g., for TiO_2.[20] Optical effects are inhomogeneities and anisotropies, i.e., complex refractive-index changes with variable film thickness and angle of incidence of light. Optical anisotropies caused by columnar film growth can be used for special applications, such as polarizers for normal incidence.[21,22] This application is the most prominent example of how to make a virtue of necessity and how a microstructure can be tailored by means of growth phenomena.

Optical absorption is caused by defects hosted in the porous film structure. In most cases these defects are water, oxygen, and hydrocarbons. As mentioned above, thin films contain higher numbers of defects, by ~5 orders of magnitude, than do bulk materials. To estimate how a concentration N_D of absorbing defects per volume unit influences absorption coefficient β, one can use the simple relation

$$\beta = \sigma N_D. \tag{3}$$

Table 1. Rate of Absorption Values to Defect Concentration in Dielectric Thin Films

Type of Film	N_D (cm^{-3})	β (cm^{-1})	$1/\beta$	$k_{250\text{ nm}}$
Single crystal	10^{13}	10^{-3}	10 m	2×10^{-9}
Thin film	10^{18}	10^{2}	100 μm	2×10^{-4}
Basic absorption	10^{22}	10^{6}	10 nm	2

Absorption cross section σ for one photon is approximated by the area of one atom (10^{-16} cm^2).[23] Table 1 lists typical defect concentrations (N_D), absorption coefficients (β), optical penetration depths ($1/\beta$), and extinction coefficients ($k_{250\text{nm}}$) at 250-nm wavelength. The photon absorption cross section was estimated to be 10^{-16} cm^2. Basic absorption means absorption for wavelengths below the shortwave absorption edge.

Furthermore, real structure causes scatter losses. All types of real-structure phenomena such as grains, pores, defects, and rough interfaces are directly connected to scattering.[24]

5. Metal Films

Growth of metal films starts with islands (the Volmer–Weber mechanism). Island films are transparent to visible and infrared radiation. At percolation thickness the film becomes continuous and is transparent in the visible spectral region while it reflects the infrared. Finally, at larger thicknesses all radiation is reflected. This process is one of the most exciting phenomena in optics: Properties change with film real structure only by addition of some nanometers of metal material, as is shown schematically in Fig. 8.

A. Metal Island Films

As has been known for many years, metal island films (Ag, Au, Cu, . . .) do not behave optically similarly to the corresponding bulk material.[25] Fifty years ago this effect was called *Anomalien der optischen Konstanten* (anomalies of optical constants). Today these properties are of outstanding importance in optics.[26] Metal islands may interact with light over surface plasmons, namely, by agency of plasmon–polaritons. Plasmons are strong collective oscillations of electrons in the island in phase with the incoming light. The Mie resonance frequency of the oscillation depends on three factors: the electronic

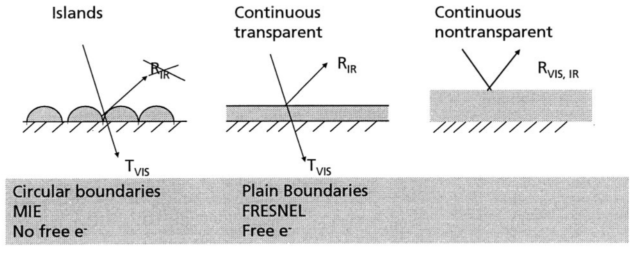


Fig. 8. Growth of a metal film, starting from islands to continuous transparent to nontransparent films.

properties of the metal, the size and shape of the islands, and the properties of host material in which the islands are embedded or upon whose surface they are fixed.

Because metals are highly absorbing, only small changes in cluster size and distribution can be used to engineer the optical properties of the ensemble. Optical constants can be calculated on the basis of Maxwell–Garnett models and of the Drude theory of free electrons.[27,28] Practical applications of island films require a macroscopic matrix in which the sensitive nanometer-sized metal clusters are embedded. In an optical film design, metal islands can be embedded in barrier layers such as SiO_x, Al_2O_3, and NiCr to prevent oxidation or in other matrices such as transparent conductive oxides. Metal island films have become important elements of present-day optics and optoelectronics. Intensive research is being conducted into active optoelectronic elements that consist of island films, including optically active individual molecules. As an example of optically active molecules, the Stranski–Krastanov layer-plus-island growth of Ge islands upon Si substrates for the production of self-assembled quantum dots may be mentioned.[29] Such deposition can be performed only under ultrahigh vacuum conditions and upon clean single crystalline surfaces. In optics, deposition conditions are far from being so well defined. However, there are many industrial applications that make use of the optical, electronic, and mechanical properties of metal island films. Classic examples of nano-optics are the red and yellow colors of Middle Ages church windows, photographic systems, and solar absorbers. Recent applications in the field of photonic bandgap materials are based on ultrafast light-switching phenomena, which can by used as novel components in integrated optics for biosensors or optical tweezers. We are just beginning to use them in the engineering of novel optical films with extraordinary properties.

B. Continuous Transparent Metal Films

At thicknesses greater than the percolation thickness, metal films behave optically similarly to the corresponding bulk materials and are transparent to visible radiation and reflective for infrared radiation as long as the thickness is less than ~20 nm. In this thickness interval, transparent metal films can be used as neutral beam splitters, induced transmission filters, solar control coatings, thermal insulating coatings, transparent electrodes, and heating layers. For example, silver is used for neutral-color highly transparent and thermal insulation coatings (low emissivity), variably colored solar control coatings, and low-emissivity Sun coatings.[30,31] For reduction of absorption, the percolation thickness should be as low as possible. Maximum wetting of the substrate can be achieved in the two-dimensional Frank–van der Merwe growth mode. Unfortunately, as was already mentioned, optical film growth starts with three-dimensional islands. In that case one can decrease the percolation thickness by increasing the number of islands per unit area. At the same mean film thickness, many small islands cover a larger substrate area than do fewer but large islands. Tailoring percolation thickness to minimum values by increasing the deposition rate and reducing the substrate temperature is not possible, owing to the severe technological restrictions that are typical for large-area architectural coatings. However, the concept of changing surface energies (Fig. 5) can be applied successfully. This can be done by plasma pretreatment of the substrate or by use of ultrathin bonding layers such as Cr, Pd, and NiCr. Also, ion assistance can be used.[32] At larger thicknesses, however, abnormal grain growth occurs,[33] as predicted by Grovenor's structure zone model[18] (Fig. 6). Wetting can also be improved by the use of such semiconductive transparent oxides as In_2O_3, SnO_2, ZnO_2, and In_2O_3 doped with 2–10% Sn. These oxides are frequently used instead of, or mixtured with, metals. Finding the best engineering technology is currently a field of keen competition among large-area coaters. State-of-the-art basic knowledge of film growth is of hardly any help today for finding solutions to improve wetting.

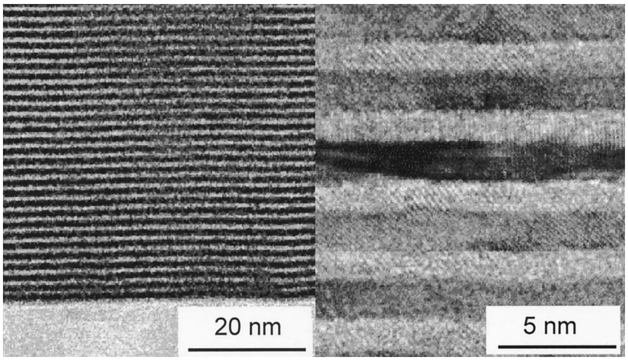


Fig. 9. Cross-sectional transmission electron microscopy picture of Cr–Sc multilayer mirrors with period spacing $d = 1.57$ nm (left) and $d = 3.17$ nm (right).[34]

Another application of continuous metal films is as multilayer mirror coatings for soft x radiation (especially in the water window: 2.3–4.4 nm) and for extreme-ultraviolet radiation (10–100 nm). Important applications in this context are microlithography, x-ray astronomy, plasma spectroscopy, medical engineering, x-ray microscopy on live tissue, and laser research and synchrotron radiation research.[34] What has been achieved is near-picometer precision for the production of nanometer multilayer mirrors for x rays (Fig. 9). With ultraprecise biased magnetron sputtering it takes only approximately an hour to deposit 600 layers for that wavelength range.[35]

C. Continuous Nontransparent Metal Films

Continuous nontransparent metal films have long been used as mirrors. It has been shown that, at high deposition rates and low substrate temperatures, nucleation density is high and that consequently thin films have a fine polycrystalline microstructure and maximum reflectivity. This has been known for many years by industrial producers of metal mirrors as the "quick-and-cold rule."

6. Summary

The growth of optical thin films starts with adsorption and diffusion of individual atoms. The structure of polished optical surfaces is rather less well-defined in terms of surface energy and can be characterized only by integral values. The maximum permissible substrate temperature in optics is ~300 °C, and deposition rates are in the ranges of some nanometers per second. At the same rate, foreign atoms from the residual gas impinge upon the substrate surface. Therefore nucleation starts statistically with the formation of three-dimensional islands (Volmer–Weber mechanism). Next, coalescence leads to a macroscopic network, and at percolation thickness the optical and also all other properties change abruptly. Further thickness growth leads often to a polycrystalline real structure. Films consist of grains that range from a few nanometers to several micrometers in size. Grain boundary regions can have volume fractions comparable with those of the grains. Consequently the properties of thin films differ markedly from the bulk properties of the same material. Thin-film properties are directly related to the film's real structure. The study of structure–property relationships is therefore indispensable for engineering thin films. Substrate temperature and deposition rate can be varied only within a limited range. Film growth can be influenced more effectively by changes of the substrate surface energy and an increase in the energy of adatoms.

Substrate surfaces can be pretreated with different kinds of plasmas and ions and by prenucleation with subnanometer bonding islands. The energy of adatoms can be increased by all types of energetic deposition, especially by use of ion assistance. Where applicable, sputter technology leads to much more defined film properties, because the energy of adatoms is in the region of some electron volts, compared to only 0.1 eV with electron-beam or boat evaporation. Film growth in optics in contrast to in electronics is rather undefined and poorly understood. Despite the fact that optical coatings are used with an ever-increasing leverage effect on optics, we are only just beginning to understand film growth in detail. Film growth is a unique low-cost nanofabrication process. It has the property of self-assembling material over large areas into periodic structures that exhibit photonic bandgap properties. For controlling the optical properties of films nowadays it is of utmost importance to have a thorough knowledge of real structure on an atomic scale.

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