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## Epitaxial growth of thin films—a statistical mechanical model

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### Abstract

A theoretical framework is developed to describe experiments on the structure of epitaxial thin films, particularly niobium on sapphire. We extend the hypothesis of dynamical scaling to apply to the structure of thin films from its conventional application to simple surfaces. We then present a phenomenological continuum theory that provides a good description of the observed scattering and the measured exponents. Finally the results of experiment and theory are compared.

Although there has been great interest in the theoretical modelling of molecular beam epitaxy (MBE) growth [1], there have been relatively few attempts to compare the detailed predictions of the models with experimental results. Also, models developed from statistical mechanics have mostly concentrated on the shape of the growing interface rather than on the structure of the grown material, whereas many experiments provide information about the whole film. Microscopic models of thin films have been developed [2] and successfully describe the structures of very thin films consisting of only a few atomic layers, but far less has been achieved in understanding the detailed structure of thicker films, 100–10 000 Å, that are of importance for many applications. In this paper, we present a dynamical scaling hypothesis for the structure of thin films involving the displacement of the atoms from their ideal positions. On the basis of this, we compare the results of x-ray scattering studies of thin films [3,4] with the predictions of a continuum model for MBE growth developed by Villain [5].

It is well known [6] that for x-ray scattering from a single interface, the relative intensity of the specular reflection yields information about the mean square width of the interface,  $\langle h(x, t)^2 \rangle$ , in the direction perpendicular to the interface, while the form of the non-specular scattering provides information about the correlation function  $\langle h(x, t)h(x + r, t) \rangle$ , where  $x$  and  $r$  are two-dimensional position vectors in the plane of the interface, and the averages are taken over all positions  $x$  on the interface. The scattering from thin films requires more discussion [7]. For a wavevector transfer  $Q$ , the scattering from a monatomic film is given by

$$I(Q) \propto \sum_{j,k} \exp(i[Q_{\parallel} \cdot (r_j - r_k) + Q_z(z_j - z_k)]) \quad (1)$$

where  $(\mathbf{r}_j, z_j)$  is the three-dimensional position of the atom  $j$ , the summations are over all the atoms in the film, and the growth direction is parallel to the  $z$ -axis. We shall consider the scattering near the Bragg reflection, with  $\mathbf{Q} = \mathbf{G}$  parallel to the growth direction of the film so that  $Q_{\parallel} = q$  and  $Q_z = G + q_z$ , with both  $q$  and  $q_z$  being small compared with the reciprocal-lattice vector  $G$ . The positions of the atoms in the film are written in terms of the atomic positions of the crystallographically perfect structure  $(\mathbf{R}_j, Z_j)$ ; the displacements from these positions for the real structure are written as  $z_j = Z_j + u_j$ . Equation (1) can then be rewritten as

$$I(\mathbf{Q}) = \sum_{j,k} \langle \exp(i[\mathbf{q} \cdot (\mathbf{R}_j - \mathbf{R}_k) + q_z(Z_j - Z_k) + G(u_j - u_k)]) \rangle \quad (2)$$

where it has been assumed that  $\mathbf{q}$  and the displacements  $u_j$  are sufficiently small that terms of second order in these variables can be neglected. The sum over the  $z$ -components is from the flat interface with the substrate  $Z = 0$  to the height of the film  $Z_j = d + h_j$ , where  $d$  is the average thickness of the film and the  $h_j$  are the fluctuations in the thickness whose average is zero. The Bragg-like component of the scattering  $I_B$  occurs for  $|\mathbf{q}| = 0$ ; if the distribution of the displacements and of the thickness fluctuations is Gaussian,  $I_B$  is given by

$$I_B(\mathbf{Q}) = D(q_z) \delta(\mathbf{q}) \exp(-G^2 \langle u^2 \rangle), \quad (3)$$

where  $D(q_z)$  is the Debye–Waller-like term arising from the fluctuations in the thickness of the film, given in turn by

$$D(q_z) = (1 - 2 \cos(q_z d) \exp(-q_z^2 \langle h^2 \rangle / 2) + \exp(-q_z^2 \langle h^2 \rangle)) / q_z^2.$$

This equation can then be further approximated to give

$$I_D(\mathbf{Q}) = 4q_z^{-2} \sin^2(q_z d / 2) \delta(\mathbf{q}) \exp(-G^2 \langle u^2 \rangle - q_z^2 \langle h^2 \rangle). \quad (4)$$

The leading term in the diffuse or non-Bragg-like scattering is obtained by expanding the exponential term in equation (2) and assuming that the displacements from the ideal positions and the fluctuations in the heights are sufficiently small that only the leading terms need to be considered. The expression for  $q_z = 0$  is then

$$I_D(\mathbf{Q}) = \sum_{j,k} \langle (d^2 G^2 \overline{u_j u_k} + h_j h_k) \exp(i\mathbf{q} \cdot (\mathbf{R}_j - \mathbf{R}_k)) \rangle. \quad (5)$$

The displacements are averaged through the thickness of the film and are written as  $\overline{u_j}$ , so the summations over  $j$  and  $k$  are now over all the lattice positions in the plane of the film.

Equations (3) and (4) generalize to a thin film the expressions for the scattering from a single interface. The thin film has a conformal structure if the fluctuations of  $\overline{u}$  are proportional to those of  $h$ . This corresponds to the case where large fluctuations in the displacements are correlated with large fluctuations in the film thickness. The two terms in the scattering can then be combined into a single term. This assumption of conformality enables us to propose an *extended dynamical scaling hypothesis for thin films*. The scattering is then solely a function of the fluctuations in the thickness/height instead of the two variables of the height and the atomic displacements. Equations (4) and (5) form the basis of the scaling hypothesis for the scattering from thin films.

With this assumption we now use conventional scaling relations for the thickness of the film (which we have argued is conformal with the atomic displacements) and their relationship to the scattered intensity, to define the critical exponents  $\alpha$  and  $\beta$ :

$$u^2(t) = \langle h(\mathbf{x}, t)^2 \rangle \approx t^{2\beta}, \quad t \ll L^z, t, L \rightarrow \infty \quad (6)$$

where  $L$  is the size of the film and  $z = \alpha/\beta$  is the dynamical critical exponent,

$$\langle h(\mathbf{x}, t) h(\mathbf{x} + \mathbf{r}, t) \rangle \approx |\mathbf{r}|^{2\alpha}, \quad |\mathbf{r}| \rightarrow \infty \quad (7)$$

where the equalities above are valid asymptotically, and the scattered intensity is the Fourier transform of equation (7):

$$I(q) = \langle h(\mathbf{q}, t)h(-\mathbf{q}, t) \rangle.$$

A continuum model of a surface was proposed by Villain [5] based on the Edwards–Wilkinson (EW) equation [8] with the addition of a surface diffusion term. The model leads to the differential equation

$$\partial h(\mathbf{x}, t)/\partial t = -B \nabla^4 h(\mathbf{x}, t) + A \nabla^2 h(\mathbf{x}, t) + \eta(\mathbf{x}, t). \quad (8)$$

The first term on the RHS models the surface diffusion, the second is the normal diffusive term, and  $\eta(\mathbf{x}, t)$  is the noise that is assumed as usual to have a Gaussian distribution. When the surface diffusion term is neglected, the equation reduces to the EW equation, which has been used as an approximation to the KPZ [9] equation to model the growth of multilayers [10]. We are unaware however of any comparison of the predictions of equation (8) with experimental data and in particular of the justification of the presence of the different terms therein in the context of a specific experiment.

In his important work on the theory of MBE growth, Villain [5] pointed out the physical relevance of the terms in equation (8). In the presence of desorption, the rate of change of  $h(\mathbf{x}, t)$  can be written as the difference between the chemical potential  $\mu$  at a point  $(\mathbf{x}, t)$  on the surface and the average chemical potential  $\mu_c$  of the atoms in the molecular beam; arguing further that the consequent growth cannot depend on the orientation of the plane leads to

$$\frac{\partial h(\mathbf{x}, t)}{\partial t} \propto (\mu(\mathbf{x}, t) - \mu_c) = B \nabla^2 h(\mathbf{x}, t). \quad (9)$$

A further source of the  $\nabla^2 h(\mathbf{x}, t)$  term has been argued to be the downward funnelling of atoms to the lowest ‘valley’ available [11] to them.

Turning to the surface diffusion term  $\nabla^4 h(\mathbf{x}, t)$ , the rate of change of  $h$  is argued [4] to obey a continuity equation with respect to a current density  $j$ , which is itself the gradient of the chemical potential  $\nabla^2 h(\mathbf{x}, t)$ :

$$\frac{\partial h(\mathbf{x}, t)}{\partial t} = -\nabla \cdot \mathbf{j}(\mathbf{x}, t) = -A \nabla^2 \mu(\mathbf{x}, t) = -A \nabla^4 h(\mathbf{x}, t). \quad (10)$$

This term is characteristic of MBE growth, and is typically known as the noiseless Wolf–Villain equation [11, 12]. It is a crucial ingredient in any serious model of MBE growth. Finally we remark that equation (8) is applicable to the motion of atoms on stepped surfaces [5]; additionally, downward funnelling, finite desorption, and surface diffusion are valid physical processes and hence equation (8) is a reasonable model for describing the experimental conditions for MBE growth.

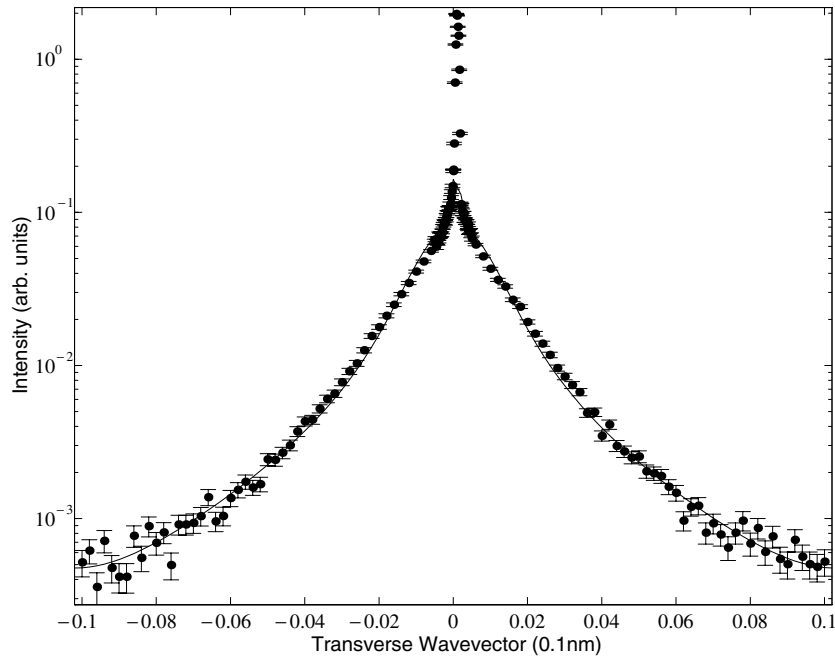
Equation (8) is linear and can be solved [5] exactly. If the system is also isotropic, the solution depends on  $q = |\mathbf{q}|$ :

$$\begin{aligned} \langle h(\mathbf{q}, t)h(-\mathbf{q}, t) \rangle &= \langle \eta(\mathbf{x}, t)\eta(\mathbf{x}', t') \rangle \{1 - \exp[-2(Aq^2 + Bq^4)t]\} / (Aq^2 + Bq^4), \\ \langle h(\mathbf{q}, t)h(-\mathbf{q}, t) \rangle &= C_0 [1 - \exp(-2\gamma(q)t)] / \gamma(q) \end{aligned} \quad (11)$$

where  $C_0$  is a constant depending on the amplitude of the noise term and  $\gamma(q) = Aq^2 + Bq^4$ . If we assume that the film thickness  $d$  is proportional to time, as is assumed in theories of kinetic roughening [1], the diffuse scattering is given by

$$I_D(q) = C_1 [1 - \exp(-2C_2\gamma(q)d)] / \gamma(q) \quad (12)$$

where this expression follows from our dynamical scaling hypothesis for thin films, and  $C_1$  and  $C_2$  are constants.



**Figure 1.** The x-ray scattering intensity from a transverse scan of the wavevector transfer through the (220) Bragg reflection from the Nb layer, 20 nm thick. The observed intensity is fitted to a Gaussian to represent the Bragg reflection and the function proposed in the text for the diffuse scattering.

Figure 1 shows the x-ray scattering observed [3] from one of the thin films of niobium grown on sapphire single-crystal substrates. The thickness of all the films is larger than the critical thickness and so the lattice parameters of the films are relaxing towards the bulk niobium lattice parameter. The solid curve shows the result of a least-squares fit of the parameters  $C_1$ ,  $C_2$ ,  $A$ , and  $B$  in equation (12) to the data and we see immediately that equation (12) provides a very satisfactory description of the diffuse scattering. In particular for large  $q$ , the intensity is inversely proportional to  $\gamma(q)$  and so describes the  $q^{-4}$ -wings of the scattering [3] very well, while at small  $q$  the theory has a finite value of the diffuse scattering as found in the experiments. Similar good agreement was obtained for the other layers and for the scattering around the (110) Bragg reflection.

The full width at half-maximum (FWHM) of the diffuse scattering in  $q$  was found [3] to vary with the layer thickness  $d$  as  $d^{-0.51 \pm 0.05}$ . Assuming as usual that thickness is proportional to time, this suggests that the FWHM varies as  $t^{-1/2}$ . The correlation length of the model is defined [1] by  $\xi(t) \sim t^{1/z}$  and the FWHM is proportional to  $1/\xi$ . The value of the exponent then implies that the behaviour of the FWHM is dominated by the diffusive,  $\nabla^2 h(x, t)$ , term in equation (8). Since the line-shape for large  $q$  requires the quartic term, these two results strongly suggest that *both* the quadratic and quartic terms in equation (8) are needed to explain the experimental results.

The experiments also measured the mean square fluctuation of the structure as a function of the thickness and found that  $u^2(d) \sim d^{0.68 \pm 0.05}$ , implying that the temporal roughening exponent is  $\beta = 0.34 \pm 0.03$ . This value of  $\beta$  is the value that is obtained if  $A = 0$  for the solution of equation (8) for a one-dimensional surface; it is the value corresponding to the Wolf–Villain model, i.e., with only the quartic term in equation (8). We therefore note that the

behaviours of  $u(d)$  and the FWHM are in agreement with respectively the  $A = 0$  (quartic term only) and  $B = 0$  solutions (quadratic term only) of equation (8) for a one-dimensional surface. Normally we would expect a two-dimensional system to exhibit logarithmic roughening [1], and we shall discuss below possible reasons for a dimensionality shift.

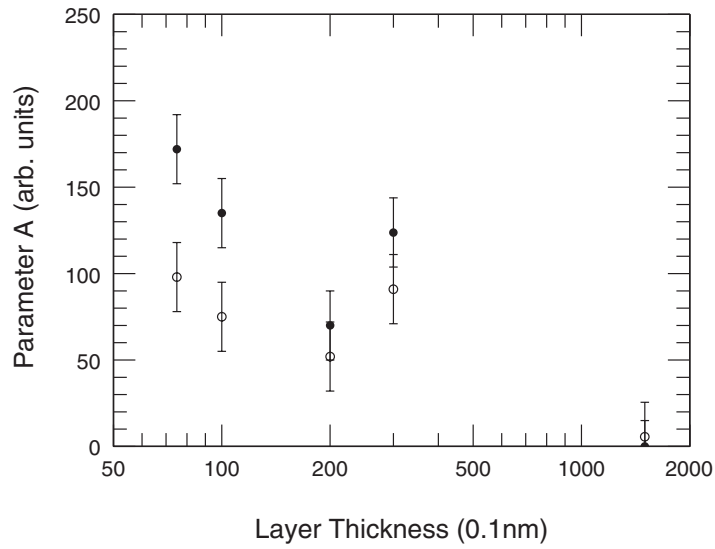
We note that the spatial roughening exponent for the one-dimensional Wolf–Villain equation, (10), is  $\alpha = 1.5$  which is an indication of anomalous scaling behaviour [1]. Since the  $\beta$ -exponent deduced from the measurements of  $u(d)$  is in good accord with the Wolf–Villain model, we consider that at least in some regime the films should exhibit anomalous scaling. There are some indications of this in figure 4 of [3]; the scattering widths as a function of  $q_x$ , i.e. perpendicular to the growth direction showed an interesting behaviour. For small  $q_x$  (and corresponding to a length scale larger than the film thickness  $d$ ), they were largely independent of  $q_x$ , but for larger  $q_x$ , they increased approximately linearly with  $q_x$ . This was earlier [3] given the interpretation that in the latter case, the displacements with these wavevectors did not propagate through the film, while in the former case, the scattering profile was characteristic of the whole thickness of the film. Another way of seeing this, however, is to say that the distribution of the atomic displacements  $\langle u_z^2 \rangle$  depends on the length scale over which it is examined. If the film is conformal, it is conceivable that this inhomogeneity in the distribution of atomic displacements should be reflected in surface fluctuations, which would then exhibit the same length scale dependence, leading in turn to anomalous scaling.

Other experimental measurements of the spatial roughening exponent in metallic materials have given values of  $\alpha = 0.79 \pm 0.05$  [14],  $0.65 \pm 0.03$  [15], and  $0.85 \pm 0.05$  [16]. These values are intermediate between the values of the exponents predicted by the one-dimensional EW equation ( $\nabla^2 h$ ), 1.5, and that of the Wolf–Villain model ( $\nabla^4 h$ ), 0.5, which are the two limiting cases of our equation (8). Further investigations are needed to determine whether the experimental systems need entirely different theoretical models to describe them, or whether they could be treated as intermediate cases that could be accommodated within the general framework of our present work.

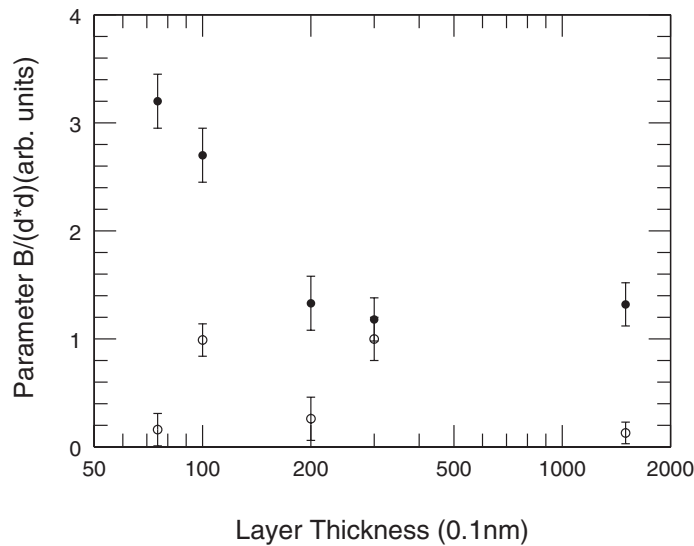
Apart from the values of the exponents, the fits to the experimental results provide information about the values of the parameters in equation (11). The measurements were made in such a way that the scattered intensity could not be reliably scaled from one sample to another and so the constants  $A$  and  $B$  were obtained by adjusting the overall scale factor to give the appropriate total scattered intensity for the thickness of the film. The results for parameter  $A$  are shown in figure 2. There is considerable scatter in the results at least partly because the parameters in the least-squares fits are strongly coupled, and both  $A$  and  $B$  are consistently larger when deduced from the (110) data than when deduced from the (220) data. As discussed in [3], some of the differences between the films may arise because they were grown at different times and capped in different ways. The difference between the results obtained from scattering near the (110) and (220) Bragg reflections may arise from a failure of the small-displacement approximation. Also, the smallness of the value of  $A$  obtained for the film with a thickness of 1500 Å may be caused by the correlation length being then so small that it is difficult to distinguish between the Bragg component and the diffuse scattering.

Figure 3 shows the thickness dependence of the  $B$ -parameter. This term arises from the quartic term in the differential equation (8) and so to compare it with the  $A$ -term we need to scale it by the square of a length. In figure 3 we show  $B/d^2$  because we found empirically that this quantity was largely independent of  $d$ .

A full microscopic theory of the structure of thin films is not yet available. The distortions from a perfect structure arise presumably from the existence of threading dislocations as explained, for example, by Kaganer et al [17]. Although their theory can be used to describe



**Figure 2.** The parameter  $A$  as a function of the layer thickness as deduced from scattering near the (110) Bragg reflection (filled points) and the (220) reflection (open points).



**Figure 3.** The parameter  $B/d^2$  as a function of layer thickness. The data were obtained from scattering near the (110) Bragg reflection (filled points) and the (220) reflection (open points).

thin films with very low dislocation densities or for very thick films, it is the intermediate region that is important for many applications.

In conclusion, we have proposed a scaling ansatz to describe the development of the structural deformations in thin films and have applied it to describe the structure of Nb thin films grown on sapphire substrates. We note that other films, both semiconductors and metals, give similar experimental results. The scattering is well described by the profile proposed by

equations (8) and (12). The exponent measured for the spatial correlation length shows that it is dominated by the diffusive interactions (quadratic term) in the phenomenological differential equation (8), while the quartic term in equation (8) is needed to explain the temporal roughening exponent. We believe that we may in this sense have provided a minimal model for explaining various quantitative features of the experiment in [3].

Normally we would expect a two-dimensional system such as that corresponding to the experiment in [3] to exhibit logarithmic roughening [1], rather than the power-law roughening indicated by our measurement of the temporal roughening exponent,  $\beta$ . Since the simplest theories state that two-dimensional surfaces are at the critical dimension for which the roughening shows logarithmic behaviour when the diffusive term is present, the power-law result is unexpected. We note that good agreement with the observed exponent is obtained, rather than for the linear one-dimensional theory presented here.

The most likely explanation for this is that since the measurements concerned were the result of synchrotron experiments, there was a high degree of anisotropy in their resolution. In particular, in the experiments [3], the experimental resolution element perpendicular to the scattering plane was much larger than in the other directions; the data integrated over the  $x$ - $y$  plane could also have contained some highly anisotropic averaging. It could also be that there is in the experiment [3] a dimensionality shift analogous to that found in the random-field model [18] due to the initial defects present in the substrate. Another remark concerns the fact that the ratio  $B/d^2$  is approximately constant (figure 3), while  $A$  decreases with increasing  $d$ , leading to increasing dominance of  $B$  for large film thicknesses. This could occur because as the thickness of the film increases, with its length always remaining constant, the approach to asymptotic behaviour in equation (8) is slowed down, as a result of which the higher-order non-linear term (whose coefficient is  $B$ , in this case) appears to win out. We hope that the relative success of our minimal model in explaining quantitative observations on the sapphire–Nb film analysed in [3] will lead to some of these issues being resolved, and to new theoretical developments in the study of thin films and multilayers, for which the thickness is in the important range between 100 and 10 000 Å.

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