Time-Invariant Structure Factor in an Epitaxial Growth Front

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It is shown that the dynamic-scaling-hypothesis description of an epitaxial growth front leads to a time-invariant structure factor in reciprocal space. This invariant structure factor is a result of having both stationary step density and step distribution during growth. The structure factor exists in the scaling regime where both the interface width and lateral correlation length grow in time according to power laws. This invariant characteristic, which manifests itself in the short-range behavior in the multilevel crystal surface, can be readily tested in a diffraction experiment.

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Recently it has been shown that the dynamical scaling approach [1-3] is an extremely fruitful tool for describing the temporal evolution of complex interface growth problems. A fundamental understanding of the microscopic aspects of the dynamics of interface growth is not only of prime interest for thin-film growth and material science but also for numerous technological applications. In the dynamical scaling approach two correlation lengths can be defined, one perpendicular to the surface, denoted by w, and another one parallel to the surface, denoted by ξ; w is also called the interface width. In the scaling regime, the two correlation lengths evolve in time as power laws: $w\sim t^{\beta}$ and $\xi\sim t^{\beta/\alpha}$, where the exponent β is related to the rate of growth of the surface and α describes its "roughness." The values of exponents α and β depend on specific models of growth. The dynamical scaling hypothesis leads to an equal-time height-height correlation of the form [1,3]

$$H(\mathbf{r},t)=\langle[h(\mathbf{r},t)-h(0,t)]^2\rangle=2[w(t)]^2 g\left(\frac{r}{\xi(t)}\right), \quad (1)$$

with $g(x)=x^{2\alpha}$ for $x\ll 1$ and $g(x)=1$ for $x\gg 1$. Here a surface atomic position is represented by the lateral coordinates, $(x,y)=\mathbf{r}$, and the vertical coordinate (along surface normal direction), $z=h(\mathbf{r},t)$.

In this Letter we show that the dynamical-scaling-hypothesis approach leads to an interesting invariant structure factor which can be readily tested in diffraction (such as the low-energy electron diffraction and the surface x-ray diffraction) experiments.

The kinematic diffraction from crystal surfaces containing stepped structures has been well understood in two extreme cases; one is the flat surface with a narrow interface width of few atomic levels [4-6], and the other is a rough surface containing an unrestricted number of atomic levels with a divergent interface width ($w\to\infty$) [7-9]. Rigorously speaking, most of these models are not suitable for the description of a growing interface that involves a large number of atomic levels but still has a finite interface width w. The diffraction theory proposed by Sinha *et al.* [10] deals with surfaces having a large w; however, it is for a continuous surface, not for a stepped surface. Furthermore, it does not address the time-dependent growth problem.

Similar to the diffraction structure factor defined in the conventional self-similar domain growth problem [11], one can define a diffraction structure factor for the present self-affine interface growth problem as

$$S(\mathbf{k}_{\parallel},k_{\perp},t)=\int d^2r\, C_{\phi}(\mathbf{r},t)\exp(i\mathbf{k}_{\parallel}\cdot\mathbf{r}), \quad (2)$$

where $\mathbf{k}_{\parallel}$ and $k_{\perp}$ are wave vectors parallel and perpendicular to the surface, respectively. The height difference function $C_{\phi}(\mathbf{r},t)$ is defined as

$$C_{\phi}(\mathbf{r},t)=\langle\exp\{i\phi[h(\mathbf{r},t)-h(0,t)]\}\rangle \approx\exp\{-\tfrac{1}{2}[\phi]^2H(\mathbf{r},t)\}, \quad (3)$$

where $\phi=k_{\perp}c$ (c is the vertical lattice spacing), and $[\phi]$ means ϕ modulo 2π such that $-\pi\le[\phi]\le\pi$. The diffraction structure factor Eq. (2) is a measure of the fundamental nonequilibrium properties of the system and is directly proportional to the scattered intensity in a diffraction experiment. The last step in Eq. (3) is obtained by treating the height difference, $[h(\mathbf{r},t)-h(0,t)]$, as a discrete stochastic Gaussian variable [7,12]. The discrete and periodic features of a crystal surface manifest themselves in the relations $C_{2m\pi}(\mathbf{r},t)=1$ and $C_{\phi+2m\pi}(\mathbf{r},t)=C_{\phi}(\mathbf{r},t)$ ($m=0,\pm1,\pm2,\ldots$). These relations differ from the result obtained from the model proposed by Sinha *et al.* [10], where the continuous surface height distribution leads to a nonperiodic expression, $C_{\phi}(\mathbf{r})=\exp[-\phi^2H(\mathbf{r})/2]$.

Combining Eq. (2) with Eqs. (3) and (1), and expanding Eq. (2) with respect to $[\phi]^2w^2[1-g(r/\xi)]$, one can rewrite the diffraction structure factor as

$$S(\mathbf{k}_{\parallel},k_{\perp},t)\sim e^{-\Delta}\delta(\mathbf{k}_{\parallel})+S_{\rm diff}(\mathbf{k}_{\parallel},k_{\perp},t), \quad (2')$$

with the diffuse structure factor

$$S_{\rm diff}(\mathbf{k}_{\parallel},k_{\perp},t) \sim\xi^2e^{-\Delta}\sum_{n=1}^{\infty}\frac{\Delta^n}{n!}\int_0^{\infty}x\,dx[1-g(x)]^nJ_0(k_{\parallel}\xi x), \quad (4)$$

where $\Delta=[\phi]^2w^2$ and $J_0(x)$ is the zeroth-order Bessel

function. The structure factor (2') contains a δ function with a coefficient $e^{-\Delta}=e^{-[\phi]^2w^2}$, showing the global (long-range) feature of the interface during growth. That is, the surface roughens microscopically but still remains flat macroscopically. In contrast, the diffuse structure factor, Eq. (4), measures the short-range roughness characterized by the atomic step density or average terrace size.

The asymptotic form of Eq. (4) as $\Delta\gg 1$ is given by (cf. Appendix)

$$S_{\rm diff}(\mathbf{k}_\parallel,k_\perp,t)\sim(\xi\Delta^{-1/2\alpha})^2F_\alpha(k_\parallel\xi\Delta^{-1/2\alpha}), \quad (5)$$

where $F_\alpha(y)=\int_0^\infty x\,dx\,e^{-x^{2\alpha}}J_0(xy)$. Equation (5) must be time invariant since it is a function of $\xi\Delta^{-1/2\alpha}=\eta[\phi]^{-1/\alpha}$, where $\eta=\xi w^{-1/\alpha}$ is a time-invariant quantity.

Practically, the δ component in Eq. (2') must be negligibly small, because $e^{-\Delta}\to 0$, as $\Delta\gg 1$. The diffraction structure factor (2') can thus be represented only by its time-invariant diffuse component,

$$S(\mathbf{k}_\parallel,k_\perp,t)\approx S_{\rm diff}(\mathbf{k}_\parallel,k_\perp,t)$$
$$\sim([\phi]^{-1/\alpha}\eta)^2F_\alpha(k_\parallel[\phi]^{-1/\alpha}\eta)\quad(\Delta\gg 1). \quad (6)$$

As shown in the Appendix, the integration in $F_\alpha(y)$ is dominated by the height-height correlation at short distances. The time-invariant nature of Eq. (6) is indeed a result of the time-invariant characteristic of the height-height correlation at short range (globally, both w and ξ still grow in time),

$$G(\mathbf{r},t)\sim 2w^2(r/\xi)^{2\alpha}=2(r/\eta)^{2\alpha}\quad(r\ll\xi). \quad (7)$$

This interesting phenomenon can be understood from a simple growth process, the random deposition with surface diffusion, studied both by Family [1] and by Edwards and Wilkinson [13]. After a transient period of initial buildup from random fluctuations of the deposition, the competition between the fluctuations and the local diffusion has reached a balance in the short range but not in the long range. The interface thus has a short-range stationary state (time independent) while a long-range steady growth (time dependent) proceeds in the form of a dynamic scaling.

The time-invariant characteristic, shown in Eq. (6), can be readily tested in a diffraction experiment. For most diffraction techniques (except light scattering), the condition $\Delta=[\phi]^2w^2\gg 1$ can be easily satisfied at the near-"out-of-phase" diffraction conditions where $[\phi]\geq 1$. (Note that the continuous surface model [10] cannot give these near-out-of-phase conditions.) For w as small as 5, $[\phi]^2w^2$ is as large as ~ 250 at $[\phi]=\pi$ and ~ 60 at $[\phi]=\pi/2$. At these conditions, the structure factor shown in Eq. (6) is most sensitive to the variation in the step distribution. The fact that Eq. (6) is time invariant implies that both step density and step distribution do not change in time.

The line shape of the structure factor, Eq. (6), has a full width at half maximum (FWHM) proportional to $[\phi]^{1/\alpha}/\eta$. Diffraction theories have shown that in the vicinity of $[\phi]\sim\pi$, the FWHM is inversely proportional to the lateral average terrace size [9]. Therefore, η is a very important quantity which measures the atomic step density at the growing surface. Figure 1 shows the characteristics of the invariant structure factor for several different values of the step density, $1/\eta$. The roughness parameter α is assumed to be 0.3.

The short-range behavior discussed above does not include the roughness associated with the "intrinsic width" discussed recently [14,15]. The intrinsic width usually has an atom-scale lateral correlation length. Such a short-range random fluctuation can only contribute a Debye-Waller-like factor to Eq. (6) and does not change our conclusions based on the line shape of the structure factor.

Experimentally, the step density can be obtained either from the measured FWHM of the diffraction line shape or from the peak intensity as a function of $[\phi]$. According to Eq. (6), $S(0,k_\perp)\propto[\phi]^{-2/\alpha}$ and FWHM $\propto[\phi]^{1/\alpha}$; i.e., both the peak intensity $[\propto S(0,k_\perp)]$ and the FWHM have the forms of power laws in $k_\perp$. The slope of the plot of $\ln[S(0,k_\perp)]$ vs $\ln|[\phi]|$ gives the value of $-2/\alpha$ while the plot of ln(FWHM) vs $\ln|[\phi]|$ shows a slope of $1/\alpha$. The roughness exponent α can therefore be extracted from these measurements.

The other growth exponent, β, cannot be obtained from Eq. (6), which is insensitive to the global surface features. However, we can show from Eq. (4) that the

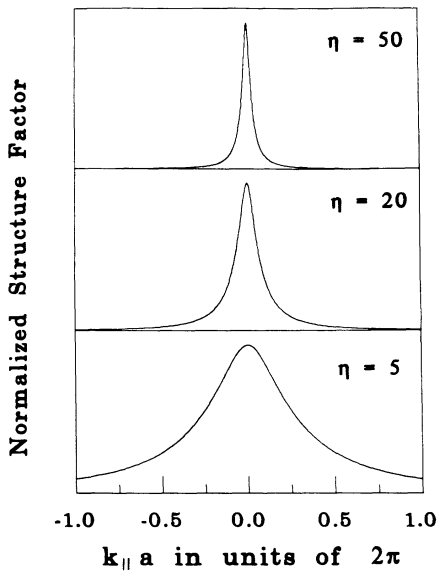

FIG. 1. The line shapes of the structure factor, Eq. (6), plotted at the out-of-phase condition, $\phi=\pi$, for different average terrace sizes, $\eta=5$, 20, and 50 (in units of the lattice constant a). The roughness exponent α is assumed to be 0.3.

structure factor (2') has the form

$$S(\mathbf{k}_\parallel,k_\perp,t)\propto e^{-\Delta}\delta(\mathbf{k}_\parallel) + e^{-\Delta}\Delta\xi^2\int_0^\infty x\,dx[1-g(x)]J_0(k_\parallel\xi x)\,, \tag{8}$$

at the near-"in-phase" diffraction condition, where $[\phi]\sim 0$ and $\Delta=[\phi]^2w^2\ll 1$. Slightly different expressions have also been obtained by Sinha *et al.* [10] and by Wollschläger, Falta, and Henzler [6] for the measurement of the interface width of a rough surface. In contrast to the behavior in Eq. (6), the structure factor at $\Delta\ll 1$, Eq. (8), is not time invariant, but is extremely sensitive to the long-range behavior of the growing interface. Experimentally, the value of β can be measured from the peak intensity as a function of time according to Eq. (8). A detailed discussion of this issue will be presented elsewhere [16].

Our results not only generalize the continuous surface diffraction theory [10] to the case of the stepped surfaces involving the time-dependent crystalline film growth problem, but also reveal several important features that have not been predicted previously. The conclusion that the out-of-phase diffraction from a multilevel crystalline interface is only sensitive to the short-range behavior provides us with a fresh insight into the surface diffraction mechanism. The time-invariant quantity $\eta=\xi w^{-1/\alpha}$, which has not been discussed before, is clearly connected to the surface step density through Eq. (6).

So far, no experimental work has been reported on the study of the time-dependent dynamic scaling behavior during the growth of a crystalline thin film. Most of the work reported to date has concentrated on the characterization of static fractal [17] properties of polycrystals [18], amorphous media, or porus media [19]. One recent experiment [20] reported a study of the dynamic scaling in an etched crystalline interface by ion bombardment. However, this is still not a direct approach for studying the common growth problems. It is hoped that our predicted invariant structure factor, which can be measured readily in a diffraction experiment, will stimulate new experimental efforts in future studies of the dynamic scaling behavior in complex nonequilibrium problems.

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Appendix: The derivation of Eq. (5).—From Eq. (4), we make a transformation, $x\leftrightarrow z$, defined by

$$e^{-z^{2\alpha}}=1-g(x)\,,\qquad x=Q(z)=g^{-1}(1-e^{-z^{2\alpha}})\,, \tag{A1}$$

where $g^{-1}(y)$ is the inverse function of $y=g(x)$. Equation (4) then becomes

$$S_{\rm diff}(\mathbf{k}_\parallel,k_\perp)\propto\xi^2\sum_{n=1}^\infty e^{-\Delta}\frac{\Delta^n}{n!}n^{-1/2\alpha}\int_0^\infty dz\,e^{-z^{2\alpha}}Q(zn^{-1/2\alpha})Q'(zn^{-1/2\alpha})J_0(S_\parallel\xi Q(zn^{-1/2\alpha}))\,. \tag{A2}$$

Since the function $Q(z)$ is expected to be an analytical function in the region $0\le z<+\infty$, one can expand the product,

$$Q(\rho)Q'(\rho)J_0(\gamma Q(\rho))=\sum_{k=0}^\infty C_k(\gamma)\rho^k\,, \tag{A3}$$

as a Taylor series in terms of ρ. Inserting Eq. (A3) into Eq. (A2) gives

$$S_{\rm diff}(\mathbf{k}_\parallel,k_\perp)\propto\xi^2\int_0^\infty dz\,e^{-z^{2\alpha}}\sum_{k=0}^\infty C_k(k_\parallel\xi)z^k\left[e^{-\Delta}\sum_{n=1}^\infty\frac{\Delta^n}{n!}n^{-(1+k)/2\alpha}\right]. \tag{A4}$$

Under the condition $\Delta\gg 1$ we can show that

$$P(\Delta,s)\equiv\sum_{n=1}^\infty\frac{\Delta^n}{n!}n^{-s}\sim e^\Delta\Delta^{-s}\quad(\Delta\gg 1)\,. \tag{A5}$$

For $s=$integer ≥ 0, the proof of Eq. (A5) is straightforward because as $\Delta\to\infty$,

$$\frac{P(\Delta,s)}{e^\Delta\Delta^{-s}}\to\frac{dP(\Delta,s)/d\Delta}{d(e^\Delta\Delta^{-s})/d\Delta}\to\frac{P(\Delta,s-1)}{e^\Delta\Delta^{-(s-1)}}\to\cdots\to\frac{P(\Delta,1)}{e^\Delta\Delta^{-1}}\to\frac{P(\Delta,0)}{e^\Delta}=\frac{e^\Delta-1}{e^\Delta}\to 1\,.$$

The rigorous proof of Eq. (A5) for an arbitrary value of s will be published elsewhere [16].

Thus, using Eq. (A5), one can simplify Eq. (A4) as

$$S_{\rm diff}(\mathbf{k}_\parallel,k_\perp)\propto\xi^2\int_0^\infty x\,dx[1-g(x)]^\Delta J_0(k_\parallel\xi x)=\int_0^\infty r\,dr\left[1-g\left(\frac{r}{\xi}\right)\right]^\Delta J_0(k_\parallel r)\,. \tag{A6}$$

The scaling function $g(x)$ indicates that

$$1-g(r/\xi)\sim\begin{cases}1-(r/\xi)^{2\alpha} & \text{for } r\ll\xi,\\ 0 & \text{for } r\gg\xi\,.\end{cases}$$

Given the condition of $\Delta \gg 1$, we have

$$\left[1-g\left(\frac{r}{\xi}\right)\right]^{\Delta}=\exp\left\{\Delta \ln\left[1-g\left(\frac{r}{\xi}\right)\right]\right\}\approx\begin{cases}e^{-\Delta(r/\xi)^{2\alpha}} & \text{for } r\ll\xi,\\ 0 & \text{for } r=\text{any other value}.\end{cases}$$

Therefore, only in the region $0\leq r\ll\xi$ can the function $[1-g(r/\xi)]^{\Delta}$ have a significant contribution to the integral of Eq. (A6). We can thus replace the function $[1-g(r/\xi)]^{\Delta}$ by $e^{-\Delta(r/\xi)^{2\alpha}}$ in Eq. (A6), which yields Eq. (5).

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[12] According to Villain, Grempel, and Lapujoulade [7], the rigorous expression of the height difference function should be

$$C_{\phi}(\mathbf{r},t)=\frac{\sum_{m=-\infty}^{m=+\infty}e^{-(1/2)H(\mathbf{r},t)(\phi-2\pi m)^2}}{\sum_{m=-\infty}^{m=+\infty}e^{-(1/2)H(\mathbf{r},t)(2\pi m)^2}},$$

where its first-order term gives Eq. (3). It can be shown that at the vicinity of $\phi\sim(2m-1)\pi$, the contributions from the higher-order terms may become significant. However, the higher-order corrections do not change the main conclusion that the structure factor is a time-invariant function at near-out-of-phase diffraction conditions. A more rigorous study on this issue will be presented in a future publication [16].

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