Faceted anomalous scaling in the epitaxial growth of semiconductor films

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Abstract – We apply the generic dynamical scaling theory (GDST) to surfaces of CdTe polycrystalline films grown in glass substrates at distinct temperatures. The analysed data were obtained with a stylus profiler having an estimated lateral resolution of 0.3 μm. We have found that the GDST applied to the surface power spectra foresees faceted morphology in the investigated temperature range. The temperature increase results in surfaces with larger faceted grains and, consequently, the higher the growth temperature the better the quantitative scaling analysis. High-resolution AFM images corroborate the morphological predictions of GDST.

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The kinetic roughening of interfaces is an outstanding topic of nonequilibrium Statistical Physics which has been intensively investigated in both theoretical [1–5] and experimental [6–10] frontlines. A generic dynamical scaling theory (GDST) includes both interface fluctuations in the real and momentum spaces [2]. The analysis in the real space can be performed using the local height-height correlation function

\[ G(l, t) = \langle [h(x + l, t) - h(x, t)]^2 \rangle, \]

where the overbar means averages over the surface profile and \( \langle \cdot \rangle \) the averages over distinct profiles. GDST gives

\[ G(l, t) = l^{2α} \Phi(l/t^{1/z}), \]

where \( α \) and \( z \) are the roughness and dynamical exponents, respectively. The scaling function \( \Phi \) behaves as

\[ \Phi(x) \sim \begin{cases} 
  x^{-2(α-α_{loc})}, & \text{if } x \ll 1, \\
  x^{-2α}, & \text{if } x \gg 1.
\end{cases} \]

The local roughness exponent \( α_{loc} \) determines the scaling locally. The power spectrum, defined as \( S(k, t) = \langle \hat{h}(k, t)\hat{h}(-k, t) \rangle \) with \( \hat{h} \) being the Fourier transform of the surface, scales as

\[ S(k, t) = k^{-(2α+d)} \Psi(k^{1/z}), \]

where the scaling function \( \Psi \) is

\[ \Psi(x) \sim \begin{cases} 
  x^{2α+d}, & \text{if } x \ll 1, \\
  x^{2(α-α_{s})}, & \text{if } x \gg 1.
\end{cases} \]

In this scaling function, \( d \) is the surface topological dimension and \( α_{s} \) is the spectral roughness exponent.

This generic scaling ansatz has been observed in a large collection of models and experiments as can be looked up in [1,6,8] and references therein. This scaling ansatz implies a constraint between the exponents that depends specially on the spectral roughness exponent [1,2,11]. If \( α_{s} < 1 \) the surface is self-affine with spectral and local roughness exponents being equal, \( α_{loc} = α_{s} \). If \( α_{s} > 1 \) the surface is locally smooth with \( α_{loc} = 1 \). Each case is still classified into two subclasses. For \( α_{s} < 1 \), if \( α_{s} = α \) we have the regular Family-Vicsek (FV) scaling, otherwise the system has intrinsically anomalous scaling. For \( α_{s} > 1 \), we have super-roughening scaling if \( α_{s} = α \) and faceted growth scaling otherwise. The constraint between \( α_{s} \) and \( α_{loc} \) is a beautiful analytical result derived from the scaling ansatz [11] while the subclasses are allusive to their physical implications [1], with the exception of the FV scaling which is due to the ansatz conceived [12].

FV, super-rough, and intrinsic scalings have been reported in several experimental works [6–8,10] but the faceted one has only been achieved quite recently in the electrodissolution of pure polycrystalline iron [6]. The surfaces undergo a transition from intrinsic to faceted anomalous scalings as dissolution time increases. The faceted regime is highly anisotropic and, consequently, the scaling analyses were performed with one-dimensional profiles, in a direction orthogonal to the anisotropy, in addition to the analyses of the two-dimensional surfaces.

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The faceted anomalous scaling was evident only for the one-dimensional case. This phenomenon was ascribed to the averages over all directions performed in $d=2$, which underestimated the scaling exponents due to contributions of unclear faceted morphology in some directions.

We investigate the anomalous roughening in semiconductor CdTe polycrystalline thick films grown on glass substrates. The real space scaling of this system was previously investigated [9,13]. In the present work, we identify a crossover in the dynamical scaling exponents not noticed in ref. [9] and perform the power spectrum scaling analysis. We find out that the GDST applied to the surface power spectra foresees faceted growth, which is confirmed by high-resolution AFM images. An inconsistency between the faceted growth presented in this work and the self-affine morphologies previously reported [9] is explained in terms of the convolution effects resulting from the finite size of the probe tip used to scan the surfaces.

The CdTe films were deposited on glass substrates covered with a transparent conducting oxide (TCO), SnO$_2$:F, resulting in a rough initial surface. The glass/TCO surface has a rms width of 16 nm that is much larger than 3 nm observed for the pure glass substrate. Although the presence of this layer introduces an undesirable large initial roughness, it is important from an application viewpoint because it is the front contact layer of solar cells produced with this material [14].

The CdTe films were deposited on glass substrates at a growth rate of approximately $1\text{nm/s}$. Details of the deposition system and sample preparation can be found elsewhere [9,15]. It is important to mention that the growth of some samples was repeated using the molecular beam epitaxy technique and they exhibited the same behaviour. The scaling analysis was done in one-dimensional profiles with 300 $\mu$m of length and 4570 pixels acquired with a stylus profiler XP1-AMBIOS. Consequently, the topological (scan) dimension is $d=1$ independently of the $(2+1)$-dimensional surface. At least 20 profiles scanned at random directions were used in the averaging.

Despite of the good vertical sensibility (better than 1 nm) and step size scan (better than 100 nm) of the profiler, the large tip radius ($\approx 1 \mu$m) limits the lateral resolution power due to convolution effects [16]. As a consequence, the data at short scales correspond to the convolution of the film surface and the probe tip geometries. A criterion accepted by the microscopy community establishes that a probe tip can resolve peaks separated up to 20% of its radius. Therefore, one can estimate, with a good margin of security, a lateral resolution $l_s = 0.3 \mu$m for this scan device. This lateral resolution, however, does not hinder the measurement of long-wavelength fluctuations and the global roughness of the entire profile can be accurately determined. The short-scale details of the surface morphology were visualized using an atomic force microscope (AFM) NTMDT-Ntegra Prima operating in the semi-contact mode. These AFM images were not included in the scaling analyses. The use of profiler scans allows to study large amplitude and wavelength fluctuations not achievable with a regular AFM. The growth time was varied from $t = 30$ to 660 min and the substrate temperatures $T = 150, 200, 250,$ and $300 \text{C}$ were analysed. The CdTe film detaches from the glass substrate and cracks during the cooling process for growth times longer than 660 min. This technical limitation hindered the growth of samples beyond this limit.

The exponents $\alpha_{loc}$ and $\alpha_s$ were obtained directly from the scaling behaviours of $G(l, t) \sim l^{2\alpha_{loc}}$ and $S(k, t) \sim k^{-(2\alpha_s + 1)}$ at $t = 660 \text{ min}$. The global interface width is defined as

$$W(t) = \left( \langle h(x, t) - \bar{h}(t) \rangle^2 \right)^{1/2} \sim t^\beta,$$

where $\beta$ is the growth exponent. GDST states $\alpha_{loc} = \min(1, \alpha_s)$ and $\alpha = \beta \varepsilon$ [2,17]. Even though the local and spectral exponents are not independent, the local roughness exponent was measured for a comparison between the theoretical prediction and experimental observation.

The surface width evolution, which is shown in fig. 1, has a crossover from a regime of low to high growth exponents. This crossover was not perceived in the former scaling analysis of this CdTe/TCO/glass system, but it was also present [9]. The crossover is probably on account of the initial surface roughness, which initially diminishes due to the occupancy of grooves and valleys. A temperature increase enhances the downward funnelling, reducing the crossover time and, consequently, the largest power-law interval was obtained for $T = 300 \text{C}$. An additional evidence for our proposition is that this crossover was not observed in the CdTe deposition on pure glass substrates (a smaller initial roughness) at the same experimental conditions [15]. Therefore, the growth exponents in ref. [9] are underestimated but the main conclusion of that work,
stating a $\beta$ exponent depending on temperature, still holds.

Whole scaling analyses were performed for the growth times after the crossovers. The $\beta$ exponents are shown in table 1. The large uncertainties for temperatures 150 and 200 $^\circ$C reflect the short time intervals of power law growth. Taking into account the uncertainties, these exponents are nearly constant for temperatures 150, 200, and 250 $^\circ$C. For $T = 300$ $^\circ$C, the growth exponent $\beta > 1$ is a signature of unstable growth that is unusually fast in the framework of kinetic roughening. However, self-affine (FV scaling) unstable growth was recently found in stochastic equations related to nonlocal interface dynamics [3,4].

Interestingly, the growth exponent obtained numerically for the unstable version of the stochastic Michelson-Sivashinsky (SMS) equation, $\beta_{\text{sms}} = 1.14$ [3], coincides with our estimate $\beta = 1.11(2)$. The number in parenthesis represents the uncertainty in the last digit meaning $\beta = 1.11 \pm 0.02$. The mentioned unstable SMS equation, written in the momentum space with suitable rescaled parameters, reads as [3]

$$\partial_t \hat{h} = (k - k^2) \hat{h} + \frac{1}{2} \mathcal{F}(|\nabla h|^2) + \hat{\eta},$$

where $\mathcal{F}(g) = \hat{g}$ is the Fourier transform of $g$ and $\eta$ a Gaussian noise. Apart from the term $k\hat{h}$, this equation is the well-known Kardar-Parisi-Zhang equation [18] related to a non-conserved local growth. The instability is due to the term $k\hat{h}$ that introduces the non-locality in the equation. However, as shown in the present work, the CdTe/Glass system is not self-affine ($\alpha_s \neq \alpha_{\text{loc}}$) and cannot be directly associated with the SMS equation.

Power spectra for $T = 300$ $^\circ$C are shown in fig. 2(a). The curves exhibit the usual plateaus for small and power law decays for large momenta. The power law yields $\alpha_s = 1.27(5)$ and, consequently, GDST states $\alpha_{\text{loc}} = 1$. Notice that momenta in a range $k < 2\pi/l_c$ were used in this analysis. Moreover, the curves shift upwards for large $k$ as time increases, implying $\alpha > \alpha_s$, since $S(k, t) \sim k^{2\alpha_s - 1 + 2(\alpha - \alpha_s)/z}$ in the scaling ansatz given by eq. (3). GDST thus predicts surfaces with anomalous scaling and facets. Even though the profiler provides coarse-grained scans that hinder to reliably resolve faceted morphologies, as can be seen in fig. 3(a), AFM images with a scan size of 5.8 nm and probe radius of 10 nm, a resolution about 100 times better than the profiler, clearly exhibit the faceted surface morphology predicted by GDST, as

| $T$ ($^\circ$C) | $\beta$ | $\alpha_s$ | $z$ | $\alpha_{\text{loc}}$ |
|----------------|--------|------------|-----|---------------------|
| 150            | 0.59(9)| –          | –   | 0.73(2)             |
| 200            | 0.56(9)| –          | –   | 0.76(3)             |
| 250            | 0.65(2)| 1.6(1)     | 3.4(4)| 0.77(2)            |
| 300            | 1.11(2)| 1.27(5)    | 1.75(5)| 0.82(2)            |

Fig. 2: (Colour on-line) (a) Power spectra of scanned profiles for $T = 300$ $^\circ$C. Growth times increase from bottom to top. The dashed line has a slope $-3.6$ and represents the scaling $S \sim k^{-1(\alpha_s + 1)}$. (b) Collapses of the power spectra using exponents $\alpha = 1.94$ and $z = 1.75$. Dashed lines represent the asymptotic behaviours of the scaling function $\Psi(x)$. The slopes are $2\alpha + 1 \approx 4.9$ and $2(\alpha - \alpha_s) \approx 1.3$.

Fig. 3: (Colour on-line) Surface morphologies for CdTe after a growth time of 300 min at a temperature of 300 $^\circ$C. (a) A 10 $\mu$m scan using the profiler. (b) A 3 $\mu$m $\times$ 3 $\mu$m AFM image illustrating the faceted CdTe surface.
shown in fig. 3(b). This is a remarkable feat of the GDST which has foreseen the faceted morphology in coars-grained data with a poor (if any) resolution of the facets. From a theoretical viewpoint, this result corroborates the claim that GDST is able to connect the scaling properties of long-wavelength fluctuations with local morphological properties.

In fig. 2(b) we probe the scaling ansatz for $\Psi(x)$, given by eq. (3), by plotting $k^{2\alpha + 1} S(k, t)$ vs. $kt^{1/2}$. The scaling relation $\alpha = \beta z$ with exponents $z = 1.75$ and $\beta = 1.11$ obtained via gradient correlation function (explained below) and global interface width (fig. 1), respectively, were used in the collapse. The excellent collapse obtained for $t > 90$ min provides an additional evidence that the data are described by anomalous scaling of faceted surfaces. Notice that the curve corresponding to $t = 30$ min, which was removed from the determination of the scaling exponents (fig. 1), does not collapse. Finally, the asymptotic scaling forms of $\Psi(x)$ are also confirmed as indicated by the dashed lines.

The dynamic exponent $z$ can be determined using the slope-slope correlation function

$$\Gamma(l, t) = \langle \nabla h(x + l, t) \nabla h(x, t) \rangle.$$  \hspace{1cm} (6)

The correlation length $\xi$, which is assumed to scale as $\xi \sim t^{1/2}$ in GDST, can be defined as the first zero of the correlation function. The correlation functions and lengths are shown in fig. 4(a). There is no clear power law regime with $\xi > l_c$ for $T = 150$–$250$ °C. For $300$ °C, the slope provides $1/z = 0.57$.

A power spectrum analysis is itself sufficient to carry out the GDST, but correlation functions are widely used in experimental investigations [6–8]. Figure 4(b) shows that the height-height correlation function, given by eq. (1), also follows the usual qualitative behaviour of anomalous scaling, in which the curves are shifted upwardly as time evolves. However, a power law regression in the linear interval indicated results in a self-affine surface with $\alpha_{loc} = 0.82(2)$, in disagreement with GDST. The scaling exponents for the other temperatures are shown in table 1. The underestimated values of $\alpha_{loc}$ are reflecting the coarse-grained resolution of the scanning device since, in this scale, the analysed data are a convolution of the surface and the probe tip morphologies. The inset of fig. 4(b) shows the correlation function collapses using the same exponents that collapsed the power spectra. These collapses are so or more convincing than those obtained for the power spectra in fig. 2(b).

The power spectra for $T = 250$ °C are shown in fig. 5(a). The curve for $t = 660$ min also exhibits a power decay in a range within $k < 2\pi/l_c$ that yields $\alpha_s = 1.6(1)$. Again, $S(k, t)$ shifts upwardly as time increases implying $\alpha > \alpha_s$. A gradient-gradient correlation function does not provide a reliable estimate of the dynamical exponent $z$ (fig. 4(a)) and thus a criterion of best collapse for $t > 90$ min was used instead to obtain $z = 3.4(4)$ and $\alpha = 2.2(2)$, as shown in fig. 5(b). Even though the scaling for $T = 250$ °C is less precise than for $T = 300$ °C, it still points out a faceted anomalous scaling which was also confirmed in AFM images. The power spectra for $T = 150$ and $200$ °C do not exhibit scaling properties accurate enough to determine the exponents but the qualitative behaviour of faceted anomalous scaling, $\alpha_s > 1$ and $\alpha > \alpha_s$, was also verified. AFM images again confirm the faceted morphologies for these temperatures. The effect of temperature is to increase the size of faceted grains and, for this reason, the scaling analysis with profiler data is more accurate at higher temperatures.

Now, we discuss the discrepancy between the $\alpha$ and $z$ exponents obtained in the present power spectrum analysis and the exponents of the real space analysis presented in ref. [13]. The first cause of difference is the underestimation of the growth exponent used to determine $\alpha$ via scaling relation $\alpha = \beta z$. The second one is the correlation length $\xi$ that overestimated the dynamical
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exponent $z$, as we clarify in this paragraph. In the previous work, the correlation length was defined as the characteristic decay in a two-point correlation function $\Gamma(l, t) = \Pr[|h(x + l) - h(x)| \leq m]$, where $m = 0.1|h_{\text{max}} - h_{\text{min}}|$ and $\Pr[A]$ is the probability that the condition $A$ is satisfied. The simple correlation function $\Gamma = \langle h(x + l)h(x) \rangle$ undergoes exactly the same effects. The correlation length $\xi$ was finally obtained by solving $\int_0^\infty \Gamma(l)dl = 0.1 \int_0^\infty \Gamma(l)dl$, where $\Gamma(l)$ is a two-exponential fit to the experimental data. However, if the tail corresponding to the long-wavelength fluctuations is or not left out of the regression, the numerical exponents may change considerably. In fig. 6, we compare the regressions of the correlation functions in two ranges: discarding (range 1) and including (range 2) the tail. The regression in the range 2, the same used in ref. [13], misfits the data only for $l \lesssim 0.3 \mu$m, whereas the regression in the range 1 fits very well the small scales but deviates for $l \gtrsim 2 \mu$m, that was the reason for the former choice of range 2. The insertion to fig. 6 shows the correlation lengths against time and the respective power law regressions. Both cases yield quite satisfactory scaling laws, $\xi_1 \sim t^{0.58}$ and $\xi_2 \sim t^{0.27}$ corresponding to ranges 1 and 2, respectively. Assuming $\xi_i \sim t^{1/z_i}$, where $i = 1, 2$, we have $z_1 = 1.72$ and $z_2 = 3.69$. Notice that the regressions provided two characteristic lengths of the same magnitude ($1 < \xi_2/\xi_1 \lesssim 2$) but exhibiting very different scaling laws with time. Repeating this analysis for lower temperatures ($T = 150–250 ^\circ \text{C}$) we found $\xi_1 \lesssim 0.5 \mu$m and no clear power law could be identified. Therefore, range 1 is the fit interval that yields the dynamical exponent consistent with GDST.

The conclusions of ref. [13] are thus partially modified since we cannot apply GDST with those exponents to infer about which dynamical scaling regime the system belongs to. The exponent $z$ presented there was misleadingly associated to the dynamical exponent of the GDST. These results also stress out the difficulties usually observed in the fitting procedure of experimental data and the problems which can arise from a misinterpretation of the parameters obtained. In this scenario, GDST in the momentum space appears as a robust method able to predict the correct scaling properties even using coarse-grained surface scans.

A renormalization group analysis of stochastic equations shows that the anomalous scaling cannot be present in local growth models [1]. Therefore, this theoretical result implies that disorder and/or nonlocal effects should be responsible for the anomalous scaling in experimental systems. Indeed, the CdTe/TCO/Glass system undergoes both effects simultaneously. The amorphous glass substrate results in random growth orientations and, consequently, a polycrystalline film is obtained. Moreover, the glass substrate and CdTe film have distinct coefficients of thermal expansion implying strained film. The interplay between nonlocal strain and substrate disorder is a possible mechanism involved in the anomalous scaling.

![Fig. 6](image-url) (Colour on-line) Height-height correlation function of the CdTe surfaces. The growth time and temperature were 90 min and 300 °C. Circles represent the experimental data while solid lines the two-exponential regressions within the indicated intervals. The inset shows the correlation length obtained for the different regression intervals.
In conclusion, we presented the scaling analysis of CdTe polycrystalline surfaces grown on glass substrates using a generic dynamical scaling theory (GDST) [2]. We investigated both height-height correlation functions and surface power spectra. The surfaces were scanned with a stylus profiler with an estimated lateral resolution of $l_c \approx 0.3 \mu m$. Surface power spectra exhibit scaling properties with a spectral roughness exponent $\alpha_s$ greater than 1 but different from the global roughness exponent $\alpha$. The GDST thus foresees faceted morphologies with local roughness exponent $\alpha_{loc} = 1$ that represents locally smooth surfaces. The height-height correlation function exhibits scaling only for $l \sim l_c$ and $\alpha_{loc}$ in the interval 0.7–0.8, indicating self-affine surfaces. However, high-resolution AFM images confirm that the surfaces are faceted. The underestimated values of $\alpha_{loc}$ are reflecting the low resolution of the scanning device since, in this range, the analyzed profile is a convolution of the surface and probe tip geometries.

It is worth to note that in a previous report on anomalous scaling in faceted morphologies [6], the surfaces are highly anisotropic and the faceted anomalous scaling was evident only for one-dimensional sections in the direction orthogonal to the anisotropy. Differently, our scaling analysis of polycrystalline CdTe consists in averaging over several randomly selected directions and, consequently, we have an experimental observation of faceted anomalous scaling in a two-dimensional analysis.

Our work opens new paths to investigate anomalous scaling experimentally. A natural extension of this work, is to check the effect of the substrate on the scaling properties. Scaling analysis of AFM images may also complement the present investigation, in particular at lower temperatures where the scaling properties did not allow to determine the scaling exponents accurately.

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