

Surface roughness and height–height correlations dependence on thickness of YBaCuO thin films

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Abstract

For high T_c superconducting multilayer applications, smooth interfaces between the individual layers are required. However, in general, e.g., YBaCuO grows in a 3D screw-dislocation or island nucleation growth mode, introducing a surface roughness. In this contribution we study the surface layer roughness as a function of different deposition techniques as well as deposition parameters. Special attention will be paid to the increase in film roughness with increasing film thickness. For these studies we used scanning probe microscopy. From these experiments, we obtained an island density decreasing with a square root dependence on the film thickness. Furthermore, height–height correlations indicate that the film growth can be described by a ballistic growth process, with very limited effective surface diffusion. The correlation lengths ξ are on the order of the island size, inferring that the island size forms the mean diffusion barrier. This results in a representation of non-correlated islands, which can be considered as autonomous systems.

Keywords: Surface roughness; Superconductors; YBaCuO thin films

1. Introduction

In order to develop a reliable device technology based on high- T_c superconducting thin films, an understanding of the basic growth processes, which play a role in the high- T_c thin film growth process, is necessary. Especially in the case of a 3D or screw dislocation mediated growth mechanism, interface roughness is an inevitable phenomenon and knowledge of the influence of the deposition conditions on the resulting morphology is an important issue.

The main goal is to characterize the kinetic roughening behaviour of single YBCO thin films as a function of thin film thickness. Therefore, we determine the surface morphology of the thin film by STM/AFM and quantify this morphology by a height–height correlation function. The obtained results are compared with the predictions of a dynamical scaling theory which in general describes the roughness evolution during thin film growth.

1.1. Height–height correlation functions

During the growth, two competing processes play a role in the evolution of the surface morphology (excluding chemical reactions). Firstly, random fluctuations occur all

the time resulting in height fluctuations and kinetic roughness build up. Secondly, smoothing effects, such as thermal diffusion or side growth, tend to eliminate the height fluctuations. These smoothing effects can only take place at a length scale smaller than or equal to the diffusion length of the add-atoms. The random fluctuations can take place on both a long as well as a short range scale. As a result, these two processes can only lead to a balancing effect on a relatively short range scale and a kinetic roughening of the film will occur.

If the conditions of the growth process are such, that the resulting interface is marginally stable and the fluctuations present are relevant, the resulting rough structure can be described in terms of self-affine fractals [1]. Applying the intrinsic scaling-properties of self-affine fractals to the statistical quantities at the growing interface leads to a dynamic scaling hypothesis [2], which implies that a growing surface is scale-invariant in both time and space or:

$$\begin{cases} r \rightarrow \lambda r & \text{(lateral scale)} \\ h \rightarrow \lambda^\alpha h & \text{(height scale)} \\ t \rightarrow \lambda^z t & \text{(time scale)} \end{cases}$$

Here, α and z represent the growth exponents and λ is the lateral rescaling factor. The exponent α is a measure for the surface roughness and the exponent z , given by

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$z = \alpha/\beta$, describes how the relaxation time depends on the system size and is called the dynamic scaling exponent. The factor β describes the time-dependent scaling of the system. We can now introduce the height-height correlation $G(r,t)$ of the growing surface, which gives the height of a surface at a position r and time t relative to the average value $\langle h(r,t) \rangle$. For G we can deduce:

$$G(r,t) \propto \begin{cases} t^{2\beta} & \text{for } r \gg t^{1/z} \\ r^{2\alpha} & \text{for } r \ll t^{1/z} \end{cases}$$

Also, we can distinguish two regimes for the interface width, separated by a crossover length $\xi(t)$ given by: $\xi(t) \propto t^{1/z} = t^{\beta/\alpha}$.

The vertical correlation length, $G(r,t)$ is a measure for the global interface width and the lateral correlation length, $\xi(t)$, is the distance over which surface fluctuations have been able to spread. Both correlation lengths evolve with time in the form of power laws. It is expected [3], that the power law (algebraic) behaviour will only be eminent for an island growth mechanism. For a step flow growth mechanism, a morphological transition to a logarithmic behaviour is predicted.

1.2. Growth models

Different growth processes lead to different values for the growth exponents α and β . However, these exponents do not depend on the specific form of the atomic interactions which result in the growth of the surface. Various growth models have been proposed to calculate the growth exponents for highly idealized systems [4,5]. In the case of random deposition, with no surface restructuring or lateral growth allowed, the columns grow according to Poisson statistics so the height of a column grows in time according to a square-root power form $h(r,t) \propto t^{1/2}$ which gives $\beta = 1/2$. The other two exponents α and z can be regarded to be equal to zero. When side growth is allowed in the growth process, voids and overhangs can develop, see, e.g., the Eden growth model [6] or the ballistic deposition model [7]. In these cases the roughening exponents are given by $\alpha = 1/2$ and $\beta = 1/3$ in one- and two-dimensional systems.

During conservative growth, a compact film is formed with surface diffusion as the primary relaxation mechanism. Predictions for the growth exponents for conservative growth for simple systems are identical to those given above, but the values of the roughness exponents may vary or even become undefined depending on the relation with other effects present during the growth process, e.g., chemical reactions or pinning centers at the interface.

2. Experimental

All YBCO thin films were grown using pulsed laser deposition (248 nm, 10 Hz, and a laser energy density of

1.2 J cm^{-2}). Samples varying in thickness from 50 to 1600 nm were grown on $12 \times 12 \text{ mm}^2$ LaAlO_3 substrates. The thicknesses of the thin films corresponded to growing times of 40 s for 50 nm films to 21 min for the 1600 nm films. The deposition temperature (760°C) and oxygen pressure have been optimized, at which, both, maximum T_c (90 K) and best surface morphology are achieved.

The surface of the thin films was characterized by ambient STM and AFM. The time between deposition and imaging was in the order of 1 to 6 days. In between, all samples were stored in a desiccator. The STM was operated at a bias voltage in the order of 800 mV and a current setpoint of 300 pA. Scan sizes were typically $1.5 \times 1.5 \mu\text{m}^2$ using 400 scanpoints in each direction, resulting in a lateral resolution of 3.75 nm.

3. Results and discussion

From each scan, height-height correlation functions were obtained. The height-height correlation at a certain distance r' was determined by averaging over the squared height differences of all scanpoints separated by that distance r' . This procedure was repeated for r' ranging from 3.75 nm to 746 nm. The latter value was restricted to half of the used scan size, to ensure enough datapoints to average. For other scan sizes the r' range changes accordingly.

In Fig. 1, a typical set of height-height correlation functions is plotted versus the correlation distance r' for a 400 nm thick sample. Scaling behaviour is only observed over one decade. In addition, the cross-over to the saturated regime is rounded. For all film thicknesses, a cross-over to constant G is present although this maximum G -value (G_{max} , the squared maximum roughness variation) can differ for different scans. For every scan, we determined: (1) the lateral correlation length ξ corresponding with the cross-over point; (2) the G value at that distance;

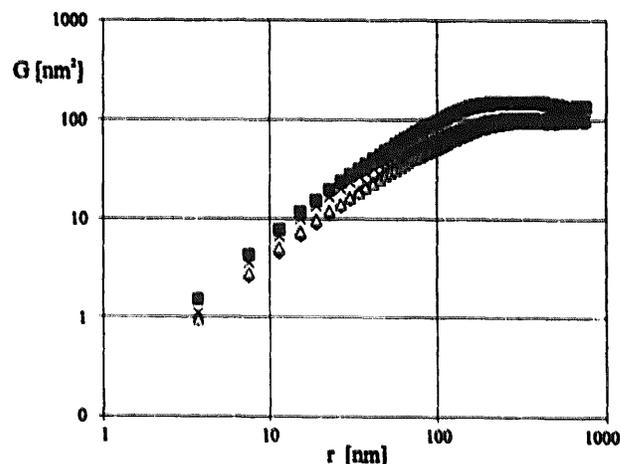


Fig. 1. Height-height correlation function $G(r)$ for four different scans on a 400 nm thick YBCO film.

(3) the averaged island size and (4) the slope in the short r' regime to determine α .

Only similar height–height correlation functions were subsequently averaged, but this was not possible in all cases. As a result, considerable variation in G_{\max} , which represents in fact the roughness variations of the thin film, is present. An initial rough substrate might result in larger G_{\max} -values, explaining this variation as an extra roughness. Since, e.g., variations in substrates are hard to account for, the lowest G_{\max} -value at a certain film thickness represents the minimum roughness obtainable. Hence, we will only use the parameters obtained from the height–height correlation corresponding with the lowest G_{\max} .

The growth exponent α as determined in the low r' regime typically over one decade and averaged over all films, is $\alpha=0.58$ with a standard deviation of 0.08. To determine the time-dependent scaling behaviour represented by the growth exponent β , we have to plot the value of $G(r')$ in the large r' regime ($r'>\xi$) versus the film thickness. $G(r)$ is then saturated and has a value G_{\max} .

We observe, within error margins, a linear relation of the G_{\max} as a function of thickness, when plotted in a double logarithmic way. The slope of the least-squares fitted curve, (exponent β) equals 0.30 ± 0.05 . Depicting the lateral correlation length ξ as a function of the film thickness in a double-logarithmic way, a linear relationship is, again, expected and the slope of the least squares fit curve should equal $1/z$ ($z=\alpha/\beta$). We find a slope of $1/z=0.43\pm 0.07$. With the previously determined α , we arrive at a $\beta=0.27\pm 0.1$. This β is close to the value obtained before. α and β values are comparable with predicted (model system) values for a ballistic deposition process with limited surface diffusion [5].

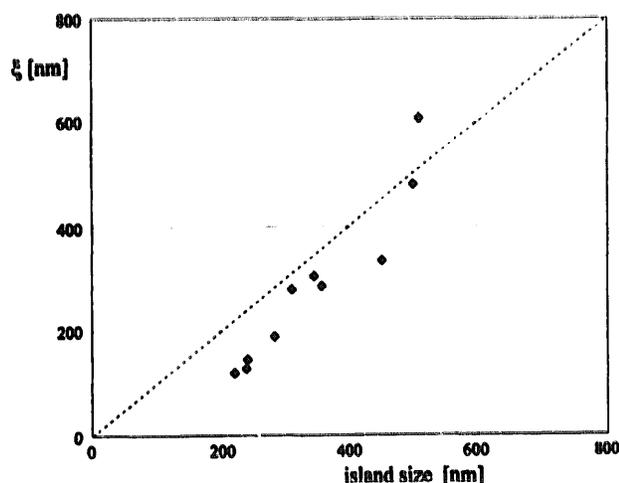


Fig. 2. Lateral cutoff point ξ versus island size.

This limiting mechanism might have its origin in the size of the islands because for all thicknesses, this size is comparable with ξ , see Fig. 2. The average size of the islands is calculated from the negative square root of the island density. Consequently, the actual island sizes will be even smaller than the ones determined from the island density, because inter-island distances are neglected.

The observation that island size and ξ -value are of the same order, indicates that correlations between atoms are limited within one island size. Hence, we can regard the system as an ensemble of independent islands. These islands increase in size with thickness and ξ increases accordingly.

4. Conclusions

With STM we examined the surface morphology evolution of superconducting c -axis oriented YBCO thin films, deposited under identical deposition conditions, as a function of film thickness in the 50 nm to 1600 nm range. To analyze the kinetic roughness behaviour, we subsequently converted the STM surface profiles into height–height correlation functions. It is expected, that these functions exhibit scaling behaviour in both time and place in the case of an island growth mechanism. We indeed find scaling behaviour in space and time resulting in growth exponents $\alpha=0.58\pm 0.08$ and $\beta=0.3\pm 0.1$. These values are comparable to theory predictions for a ballistic deposition process with limited surface diffusion, although care must be taken with theoretical comparisons, due to the complexity of the YBCO growth process and the restriction of theoretical predictions to model systems. We find a limiting mechanism, which restrains the spread of the correlations. As a result, lateral scaling behaviour is only observed over one decade. The island size and lateral correlation length ξ are of the same order, indicating that correlations between atoms are limited within one island size.

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