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Growth of quantum dots on pit-patterns

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ABSTRACT

We investigate the growth and self-organization of hetero-epitaxial quantum dots (QDs) on different patterned substrates and show that it may lead to uniform arrays of dots with different localizations. We consider the surface instability enforced by elasticity and capillarity with the wavelength λ_{ATG} and study here the influence of the pattern shape. Starting with the results for a generic sinusoidal pattern, we investigate more realistic Gaussian-like pattern shapes. Even if the evolution is intrinsically non-linear, and thence not described by the superposition principle, we show that for a pit-like pattern, the QD morphologies may be rationalized by the results of the generic sinusoidal profile with appropriate length scales. The growth of islands both on the pattern peaks or valleys is shown to result from the racing between the wetting and buried elastic dipole interactions.

1. Introduction

Si-based quantum physics leads to increasingly new developments thanks to the enhanced control over the growth of nanostructures. Controlling the properties of an assembly of such structures over large scale is a prerequisite for their use in electronic and photonic systems. Quantum dots (QDs) nucleated on strained hetero-epitaxial films usually display a significant and detrimental size dispersion and spatial disorder resulting from their stochastic origin. Similarly, self-organization may dictate their growth when instability initially leads to a periodic corrugation [1–3], but it also eventually leads to disordered dots after some coarsening.

To minimize disorder, QDs were grown on top of patterned substrates and lead to contradictory results either on top or in the bottom of the patterning. If the experiments on mesas initially found QDs sitting on the pattern tops [4,5], a significant amount of literature reported a preferential nucleation in the pattern valleys or pits [6–10], which was interpreted by means of equilibrium calculations or by kinetic barrier calculations [11,12]. Yet, initially ordered QD arrays were achieved in Ref. [13] with dots inside pits or in between pits depending on the temperature. A recent work [14] found dense and uniform QD arrays sitting on top of the pattern ‘crowns’, as initially found [4,5]. Up to now, these contradictory results were not rationalized by a unifying theoretical analysis. We use here a dynamical model which describes long-range elastic interactions and the influence of a pattern that was developed in Ref. [15]. When one considers first a basic sinusoidal pattern which induces an ‘external’ forcing with a single characteristic length, one finds a kinetic phase diagram (KPD) showing different growth regimes (where islands are found to grow either on the peaks or valleys of the pattern) as a function of the film thickness and of the ratio of the instability to the pattern wavelengths. We consider here a more general pit-pattern amenable to describe the growth on usual experimental pattern shapes. Compared to the previous sinusoidal analysis, the Gaussian-like pit profile adds an extra parameter in the phase space, the width of the pits, which has to be compared with the natural instability wavelength. We find that the results on the sinusoidal pattern may be used to rationalize the growth on such pit-patterns even if the analysis is purely non-linear.

2. Model

Quantum dots on strained hetero-epitaxial films grow by surface diffusion thanks to their elastic relaxation. In the so-called instability regime, the dots self-organize following a morphological instability [16,17] and not nucleation [3]. It was demonstrated in SiGe systems when a nucleationless evolution occurs in low-strained ($x \lesssim 0.4$) $Si_{1-x}Ge_x$ films on Si(001) [1,2]. The instability occurs only when the film is thicker than a given critical thickness H_c, and eventually leads to islands once a wetting layer and facet arise [3]. To investigate the influence of a pattern on this self-organization, we use a model which includes the effects of a modulated film/substrate interface, see Ref. [15]. The system is a crystal film in coherent epitaxy on a semi-infinite patterned substrate. Surface diffusion on top of the film free surface located at $z = h(\mathbf{r})$ with $\mathbf{r} = (x, y)$ is described by

$$\frac{\partial h}{\partial h} = D\Delta\mu, \tag{1}$$

with the diffusion coefficient D and the surface chemical potential $\mu = \delta\left(\mathcal{F}^s, +, \mathcal{F}^{el}\right)/\delta h$ where $\mathcal{F}^{el}$ is the elastic energy and $\mathcal{F}^s = \int d^2S\gamma(h, \mathbf{n})$, the surface energy. The dependence of the surface energy γ on

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both h and surface local orientation $\mathbf{n}$ depicts respectively the effects of wetting and crystalline anisotropy [18,19]. Wetting may be described by a smooth variation of γ over a few atomic lengths [3] and enforces the stable growth of 2D films below some critical thickness H_c. Anisotropy is dedicated here to SiGe systems and describes stiff (105) orientations revealed in experiments [1]. The typical elastic energy density is $\overline{\mathcal{E}}_0 = 4Ym^2(1+\nu)/(1-\nu)$ with the Young's modulus Y, Poisson's ratio ν and misfit m so that the characteristic length and instability wavelength are

$$l_0 = \frac{\gamma_f}{\overline{\mathcal{E}}_0} \quad , \quad \lambda_{\mathrm{ATG}} = \frac{8\pi}{3} l_0 , \tag{2}$$

with the film typical surface energy γ_f [16,17,3]. Thanks to the spectral analysis of the images by atomic force microscopy of the instability initial evolution, the instability length scale l_0 may be valued to typically $l_0 = 27$ nm on a $Si_{0.7}Ge_{0.3}$ film on Si(001) [20]. The film/substrate interface is located at $z = \eta(\mathbf{r})$. For a given morphology $\eta(\mathbf{r})$and $h(\mathbf{r})$, the elastic field was exactly computed in the small slope approximation as a series of the interface slopes in [15]. It involves the usual Hilbert operators associated with elasticity [21,22] together with a damping operator associated with the damping of the elastic interactions generated at the buried film/substrate interface. This damping is exponential in the film thickness (it evolves as $e^{-k\overline{H}}$ where k is a wave-vector), which introduces an explicit dependence on this thickness (contrarily to the flat substrate case, see e.g. [18]). In addition, in order to investigate the long time dynamics of the system, one needs to account for non-linear terms in the surface evolution, which involves products of Hilbert operators and of damped Hilbert operators [15]. With this solution in hand, one might numerically solve the dynamical Eq. (1) even on large scales. We consider a typical growth where the film initially follows the substrate $h(\mathbf{r}, t=0) = \eta(\mathbf{r})$, while deposition occurs up to $\bar{t}$ for a given thickness $\overline{H}$ followed by a long term annealing. (A constant F term and a deposition noise are added in the right hand side of Eq. (1) during deposition.) Capillarity initially leads to the formation of a wetting layer and for films thicker than H_c, islands emerge, grow, sometimes partially coarsen and develop facetted shapes (typically (105) pyramids). Similar to the unpatterned case, they eventually reach a stationary state without noticeable evolution (called stationary state below) after ~100 t_0 (7 h for $x = 0.3$ [20]) with the instability timescale $t_0 = l_0^4/D\gamma_f$ [23,24]. The results given below concern precisely this stationary state.

3. Harmonic analysis

We first give the results concerning the generic sinusoidal pattern

$$\eta(\mathbf{r}) = A\left[cos(2\pi x/\lambda_\eta) + cos(2\pi y/\lambda_\eta)\right] , \tag{3}$$

characterized by a single wavelength $\lambda_\eta = 2\pi/k_\eta$.

The island positions in the stationary state are dependent on two parameters

$$\lambda^* = \lambda_\eta/\lambda_{\mathrm{ATG}} \quad \text{and} \quad h^* = \bar{H}/H_c , \tag{4}$$

ruling the growth rate of the initial corrugation, and the wetting and elastic interactions. The island positioning in the stationary state is summarized in the kinetic phase diagram in Fig. 1. We find regimes where the islands form periodic arrays following the substrate geometry, either in the valleys of the pattern (label 'bottom'), or on its peaks ('top'). This occurs either when λ^*~1 (bottom/top) and/or for thin enough films (top). This organization follows either (i) a one-to-one correspondence (one island per unit cell of the pattern) where ordering is optimal, (ii) a less-than-one correspondence where coarsening (before its interruption) has led to the disappearance of some islands, and (iii) a more-than-one correspondence with a few islands on top of the pattern peaks ('clusters on top'). Particularly in (i), the island size

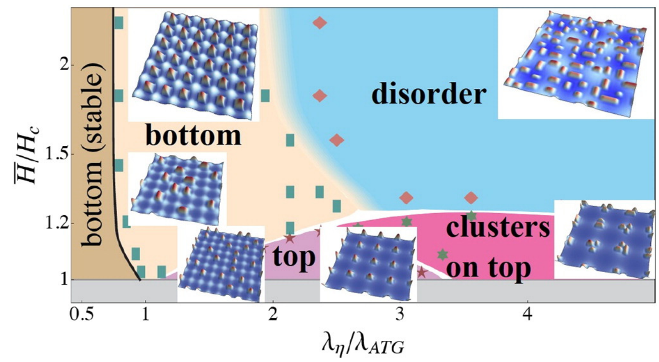

Fig. 1. Kinetic phase diagram for the position of islands grown on a sinusoidal pattern as a function of the film thickness $\overline{H}$ (in units of the critical height H_c) and of ratio of the pattern to the instability wavelengths $\lambda_\eta/\lambda_{ATG}$. The instability is inhibited on the left of the solid line and when $\overline{H}/H_c$<1.

distribution is narrow and significantly improved compared to the flat substrate case. For out-of-tuned systems (large λ^*) and/or thick enough films, the influence of the pattern is negligible and the islands self-organize with a low order (label 'disorder'), reminiscent of the growth on a flat substrate.

4. Pit-patterned substrate

The results for the generic sinusoidal pattern prove to be generic and to rationalize the island localization on more general patterns. Indeed, we consider the gaussian-like pattern

$$\eta(\mathbf{r}) = -A\, exp\left\{-\left[cos(2\pi x/\lambda_\eta) + cos(2\pi y/\lambda_\eta) + 2\right]/w\right\} , \tag{5}$$

which can mimic different geometries with the use of two characteristic length scales, the pattern period λ_η and width w. In particular, it can generate the common pit-like geometry under study in many experimental setups. Note that the following results are stable with respect to the addition of a small noise to $\eta(\mathbf{r})$. To exemplify the connection between the pit-like and the sinusoidal patterns, we give in the following, typical examples of such a connection. We first consider cases where $\lambda^* = 1.4$ and $w^* = 1$, see the results in Fig. 2. For $h^* = 1.1$, we find that the islands arise on the terraces in between the pits, stay on this position during annealing and reach a non-evolving state. For $h^* = 1.8$ however, the islands arise initially both on the terraces and in the pits. These islands then coarsen and the islands in the pits, which have a lower chemical potential, drag matter from the islands on terraces. Eventually, a well ordered and uniform array of pyramidal islands in the pits results from this growth process, and no longer coarsen. We find that these two results are in fact similar to the evolution and localization of islands grown on a sinusoidal pattern given in Fig. 1 for $\lambda^* = 1.4$ and $h^* = 1.1$ and 1.8 respectively, which will be rationalized below.

We also characterize the influence of the pit width w. We consider the case $\lambda^* = 6$, $h^* = 1.02$ for $w = 1$ and 2, see Fig. 3. When $w = 2$, the first islands arise on the terraces around the pits, and only rectangle islands remain around the pits after a long time annealing, see Fig. 3 (a–c). This result is similar to the localization on top of the pattern peaks on a sinusoidal substrate for small thickness ($h^* = 1.02$ here) and a horizontal length scale larger than ~1.2, see Fig. 1, which is the case here for both the characteristic horizontal length scales $\lambda^* = 6$ and $w = 2$. On the contrary, for $w^* = 1$, the islands directly form in the pits, coarsen and form a uniform array of pyramids in the pits, see Fig. 3(d–f). In this case, even if λ^* is large, $w^* = 1$ so that the pattern shape (and thence the film initial condition) involves in Fourier space a significant amplitude of the k_{ATG} mode. As explained below, this mode which has the largest growth