Size-selection of nanoclusters in transition from 2D to 3D growth in ion beam assisted deposition

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Abstract. Recently it has been demonstrated that by using the ion beam assisted deposition (IBAD) the cluster size selection in growth can be controlled. Particularly interesting is the Ge/Si system where IBAD leads to enhanced transition from two-dimensional (2D) to three-dimensional (3D) growth clusters, resulting to narrowing of island size distribution as compared with conventional deposition methods. In this report, size selection of nanoclusters in IBAD is studied by using a Fokker-Planck type growth model, where growth rates depend on the cluster size through the formation energy of clusters. We find that such system in IBAD may become metastable against growth with a result of narrow size distributions.

1. Introduction

Recently it has been demonstrated that ion beam assisted deposition (IBAD) can be used to facilitate self assembled Ge quantum dots in Ge/Si heteroepitaxial system and that by using pulsed IBAD it is possible to obtain narrowing of cluster size distribution as compared with conventional MBE experiments \cite{2, 1}. Moreover, the relative dispersion of the distribution in continuous IBAD is considerably larger than in MBE, while in pulsed IBAD the narrowest distributions are reached \cite{1, 2}. The effects of IBAD can be attributed to the effective lowering of the energy barriers needed to promote 2D to 3D clusters transformations. This effect can be described directly in terms of the modified energetics, in form of lowering of the transformation energy originating from the strain energy which becomes lowered by the strain relaxation due to ion bombardment. We discuss possible generic mechanisms, which may explain the size selection, and narrowing in the pulsed IBAD. The size selection of nanodots during the ion bombardment is studied by using a Fokker-Planck type model of growth, previously used successfully to explain size-selection in the kinetically determined metastable states \cite{4, 5}. Such a model can be based on the mesoscopic reaction kinetic model, where reaction rates are related to size dependent formation energy of clusters \cite{3}. In case of IBAD the basic feature of the energetics is a double-well type of energy landscape, where the wells correspond the lower energy and smaller sizes created by IBAD and the equilibrium structures, respectively. We demonstrate here that such system may become metastable against cluster growth and it thus becomes possible to reach the spontaneously selected metastable state with a narrow size distribution.
2. The phenomenological model of size selection in IBAD

In modeling the size selection in IBAD we take into account only adatom attachment and detachment processes which are related to the free energy difference $\Delta(s)$ of the formation of clusters. From this basic reaction kinetic model it is possible to derive a Fokker-Planck model of growth in form [6] (also compare with ref. with [4]) for size distribution $n(s,t)$

$$\frac{\partial n(s,t)}{\partial t} = -\frac{\partial}{\partial s}[v(s,t)n(s,t)] - \frac{\partial}{\partial x}[D(s,t)n(s,t)],$$

(1)

The time and size dependent drift and diffusion coefficients $v(s,t)$ and $D(s,t)$ are related to the reaction rates. The diffusion coefficient is simply related to the geometry of the cluster boundary and is given by $D(s) = s^q$, where $q = 1/3$ for 3D clusters. The drift velocity is given approximately by (for details of the derivation, see ref. [6])

$$v(s,t) \approx F + s^{q-1} \left[ g \frac{\Delta'(\tilde{s})}{\Delta'(\tilde{s})} + \tilde{s} \frac{\Delta''(\tilde{s})}{2 \Delta'(\tilde{s})} \right]$$

(2)

where $F$ is the adatom flux from wetting layer and/or ion bombardment induced detachments. In ordinary MBE size selection proceeds through spontaneous relaxation of predeposited layers, and $F = 0$ should be used [4]. In case of continuous IBAD there is a contribution of surface bombardment generated flux of adatoms given typically by $F \approx 0.01$ [1]. In what follows, we use dimensionless units throughout, $F \rightarrow F/(1 \text{ ML/s})$. For pulsed IBAD the average $F$ is much smaller, because between pulsing periods most surface adatoms recombine with surface defects. Therefore, the scales of interest are $F \approx 0.01$ for continuous IBAD, $F \neq 0$ but $F << 0.01$ for pulsed IBAD, and $F = 0$ for MBE. Finally, $g$ is the skewness of the size distribution $n(s)$, because size distributions have always a negative skewness (biased towards small cluster sizes) when approaching the metastable state [6].

The energetics of the model in the phenomenological level can be expressed in form a [7, 8]

$$\Delta_s = w_0 + c s^\alpha + a s^{-p},$$

(3)

where the first size-dependent term describes the "renormalized self-energy" of dots, including strain energy, while the latter term describes the coverage-dependent dot-dot interaction mediated through elastic strain fields [7]. The parameter $w_0$ sets the minimum value $\Delta_{min}$. The free energy difference $\Delta(s)$ is naturally interpreted as a one particle potential energy i.e chemical potential. Without loss of generality we can choose $q = p = 1/3$ for 3D clusters, thus reducing the number of parameters of our model. With these choices, the minimum $\Delta_{min}$ occurs at size $s_{min} = a/c$ and we redefine (setting $a = 1$) the energy and temperature scales as $\Delta \rightarrow a\Delta$ and $T \rightarrow k_B T/(\Delta_{min})$, which is dimensionless. The main effect of IBAD is to reduce the self energy by facilitating the strain relaxation, which is accounted by increasing $c$ and decreasing $\alpha$, thus reducing $s_{min}$. The total energetics of growth is now defined by allowing both the reaction paths corresponding relaxed (IBAD)and unrelaxed (MBE) states to operate in parallel, described by a double-well potential given by

$$\frac{1}{\Delta} = \frac{1}{\Delta_{MBE}} + \frac{1}{\Delta_{IBAD}},$$

(4)

where $\Delta_{MBE}$ and $\Delta_{IBAD}$ are both given by Eq. 3 but with different parameters $c$ and $\alpha$. In Fig. 1 is shown the double-well potential used in what follows and the corresponding drift velocity $v(s)$. Note that all quantities $\Delta, v$ and $F$ are now dimensionless.

The solution of the FPM can be now reduced to the Gaussian-distribution (the deviation in level of skewness already taken into account in Eq. 2)

$$n_G(s,t) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[-\frac{(s - \tilde{s})^2}{2\sigma^2}\right],$$

(5)
Figure 1. Free energy difference (dimensionless) as double-well potential (left) and corresponding drift velocity (right). The double-well potential shown for $\alpha_{IBAD} = 0.01$, $\alpha_{MBE} = 0.33$ with $q = 1/3$ (solid line) and $q = 1/2$ (dashed). The drift is shown for skewness $g = -0.1$ (solid), -0.5 (dashed) and -0.8 (dot-dashed).

where the average position $\bar{s} = \bar{s}(t)$ and the distribution width (standard deviation) $\sigma = \sigma(t)$ both depend on time and are obtained form $d\bar{s}/dt = v(s, t)$ and $d\sigma/dt = D - \beta D\Delta'(\bar{s})\sigma$ [6]. The solution of stationary average size $s_{stat}$ is obtained only numerically, but the stationary width in the metastable state is always given by temperature dependent relation

$$\sigma_{stat} = 1/\sqrt{\Delta'(\bar{s}_{stat})/k_B T}.$$  \hspace{1cm} (6)

3. Results

The approximate solutions for $\bar{s}$ and $\sigma$ are obtained in principle in analytical form from Eqs. 2, 5 and 6, but full expressions are cumbersome. In Fig. 1 we have the drift velocity for the double-well potential. For large enough sizes the effective drift becomes always negative (towards spontaneously obtained stationary state), which means that there exists a flux $F \neq 0$ which can make the distributions stationary. The barrier between the potential minima causes the distribution to broaden when the growth proceeds towards the saddle point separating the barriers. With external fluxes in region $0.001 < F < 0.01$ the distribution reaches the metastable state, and there exists a optimum drift where size selection takes place and where dispersion $\sigma/\bar{s}$ of the distribution is at optimum. In Fig. 2 the relative size $x = \bar{s}/s_{min}$ and the dispersion $d = \sigma/s_{min}$ are shown for skewed distributions at three different temperatures $T = 0.01, 0.02, 0.03$, which for the typical strain energy/particle of order 50 meV [3] correspond roughly 200, 400 and 600 K, respectively. Overcoming the spontaneously reached stationary position requires thus large external drift due to the ion bombardment, ultimately leading to formation of bi-modal distributions, but this effect is beyond the current model.

The results show that for the model potential of the type given in Eq. 3 and with selection of parameters $c$ so that minima occur at $s = 500$ and $10^4$ (corresponding the base sizes of clusters found in pulsed IBAD and MBE in ref. [1, 2], and with $\alpha = 0.33$ for MBE (see [7, 3]) and small value $\alpha = 0.01$ for IBAD corresponding nearly complete strain energy relaxation. The average stationary size $s_{stat}$ is about $s_{stat} \approx 2s_{min}$ and it does not change much at low enough values of $F$. The optimum stationary size, reached at the inflection points seen in the Fig. 2 correspond also small dispersion of order 0.1. The broadening of the distribution starts at fluxes $F \approx 0.004$ so that increasing the duration or strenght of ion bombardment does not improve the size selection but makes the dispersion of the sizes larger, and for larger fluxes, dispersion increases steadily reaching values 0.3-0.5 at $F \approx 0.1$ corresponding continuous IBAD.
Figure 2. The relative size $x = \bar{s}/s_{min}$ (left) and the dispersion $d = \sigma/s_{min}$ (right) shown for skewed distributions with $g = -0.5$ at three different temperatures $T = 0.01, 0.02, 0.03$.

in temperature region $0.01 < \tilde{T} < 0.03$. In this region $\bar{s}_{stat}/s_{min} \approx 12 - 15$, corresponding the sizes where saddle point is located. These values agree well with the experimental findings reported in ref. [1, 2], where average size $s_{puls}$ in pulsed IBAD relative to $s_{cont}$ in continuous IBAD is $s_{cont}/s_{puls} \approx (L_C/L_P)^3 \approx 23$, where $L$ is the base size reported in ref. [1, 2]. Similarly, the dispersions agree well with 10% and 30% reported for pulsed and continuous IBAD [1, 2].

4. Conclusions
We have discussed here a simple model for the size selection of nanoclusters in shape transition from 2D to 3D clusters as induced by IBAD. The model is based on the Fokker-Planck type description of the size selection in metastable state in the energy landscape which have a character of double-well potential, one of the wells describing the unrelaxed clusters with higher transformation energies and the other corresponding clusters with relaxed energies due to IBAD. The model describes how the optimum stationary size and small dispersion of order 0.1 are reached at small enough IBAD induced surface adatom fluxes $F \approx 0.004$. Increasing the duration or strenght of ion bombardment does not improve the size selection but makes the dispersion of the sizes larger. These values agree well with the experimental findings [1, 2] and suggest that size selection promoted by IBAD is related to metastable states originating from stress relaxation and its energetics induced by IBAD.

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