SAND--90-2884C

DE91 004511

DEC 05 1990

CONF 901105 25

KINETICS OF MOLECULAR BEAM EPITAXY: EFFECT OF ION-INDUCED SPUTTERING

PETER M. RICHARDS

Sandia National Laboratories, Albuquerque, New Mexico 87185-5800

ABSTRACT

Steady state roughness of surfaces growing by molecular beam epitaxy is investigated by Monte Carlo simulations under conditions where an ion beam is also present which sputters adatoms off the surface. If the sputtering is random, it only increases the roughness. But if the sputtering probability is strongly dependent on the binding energy of an adatom within a cluster or island, the ions can have a smoothening effect. Physical arguments are given in support of the results.

INTRODUCTION

Bombardment with ions such as Ar has been employed [1,2] as a possible means of smoothening surfaces either during or after deposition by molecular beam epitaxy (MBE). In some instances dramatic effects have been seen. Although the technological implications are clear, the precise mechanisms which lead to ion-induced smoothening are not well understood. This work focuses on the role played by sputtering alone. Inclusion of other ion-induced processes such as enhanced mobility and creation of adatom-vacancy complexes is likely necessary for a complete description. But the present treatment allows one to isolate one important component whose effects are not obvious. Further it is known [3] that incident ions produce substantial (~0.5 events per incident ion) sputtering. Previous simulation studies [4] have included both ion-enhanced mobility and desorption.

The simulation data and qualitative arguments given below show that random sputtering does not smoothen a rough surface; but smoothening can occur if the sputtering probability decreases strongly with the degree of bonding between the sputtered atom and its surface neighbors.

MODEL AND THEORY

I have treated the model of step-height-independent hopping in previous works [5-7] to which the reader is referred for background details and references. Those studies, however, did not consider the possibility of particles leaving the surface either by ion-induced or thermal processes. Adatoms were deposited at random and then allowed to hop with rates which both neglected interactions with other adatoms [5,6] and included strong cohesive interactions which stabilized small clusters or islands [7]. Attention was largely focused on the steady state value of the neighbor fluctuation $\sigma_1^2 = \langle (n_1 - n_2)^2 \rangle$ where n_1 and n_2 are respective heights of columns of atoms at nearest neighbor sites 1 and 2 on the substrate. One can show that, within the context of the model, the condition for steady state when random removal is included is simply

MASTER

$$R_p + R_i = 2\langle \Omega(n_j)(n_1 - n_2 - 1)\theta(n_1 - n_2) \rangle \quad (1)$$

where R_p and R_i are respective rates for random deposition ($n \rightarrow n + 1$) and removal or sputtering ($n \rightarrow n - 1$) of particles. Thus the deposition and removal processes are simply additive in regards to the roughness-producing driving term on the left-hand-side. This term is balanced by the hopping term on the right-hand-side in which $\theta(k)$ is a function which is unity for integer $k \geq 1$ and zero otherwise and $\Omega(n_j)$ is the rate for a particle to hop from site 1 to 2 thereby changing n_1 to $n_1 - 1$ and n_2 to $n_2 + 1$. If interactions are neglected, $\Omega(n_j)$ is simply a constant rate W independent of the values n_j ($j \neq 1$ or 2) of neighboring column heights. Henceforth W is taken as unity so that R_p and R_i are respective dimensionless growth and sputtering parameters. Inclusion of interactions makes $\Omega(n_j)$ depend on the environment. The particular model of ref. 7 had $\Omega(n_j) = (1 - \gamma)^{n_c}$ where n_c is the number of sites to which the particle at site 1 which makes the jump is "clustered" and γ is an interaction parameter between zero (noninteracting) and one (infinitely strong binding). A particle at site 1 is said to be clustered to neighbor site j if $n_j \geq n_1$. One should note that this model assumes the rate depends only on the initial configuration and neglects any possible dependence on the final state or details of the bonding. Consequently, for strong interactions, the diffusion rate for a "vacancy" is much less than that for an adatom, since the former requires a strongly clustered atom to jump into the hole. This is in apparent contradiction to ref. 3 where roughly equal adatom and vacancy diffusion rates were claimed. In the noninteracting case, $\Omega(n_j) = 1$, Eq. (1) can be shown to reduce to

$$R_p + R_i = \langle |n_1 - n_2| \rangle - 1 + \rho(0) \quad (2)$$

where $\rho(0)$ is the probability that neighbor columns have the same height ($n_1 = n_2$). For $R_i = 0$, Eqs. (1) and (2) are the same as found in refs. 5 and 6. The following section contains qualitative discussion based partly on Eqs. (1) and (2) together with Monte Carlo simulation results for various situations. In all cases the surfaces are taken as perfect unstepped crystal planes in the absence of deposition or sputtering. The more realistic case of vicinal surfaces is not treated here.

RESULTS FOR VARIOUS CONDITIONS OF SPUTTERING AND INTERACTIONS

Very smooth surfaces

In the limit of very low deposition and sputtering rates the situation is as depicted in Fig. 1a. There are only isolated adatoms produced by growth and "holes" produced by sputtering on an otherwise perfect surface. Mobile adatoms and holes annihilate upon meeting, thus leading to ion-induced smoothening. Corresponding simulation results are shown in Fig. 1b for the cases of deposition only, sputtering only and deposition plus sputtering. Interactions are neglected so that adatoms and holes have the same mobility. When growth and sputtering occur at equal rates, the well-known oscillations of roughness (concomitant with RHEED oscillations) for either deposition or sputtering alone are replaced by a nearly constant σ_1^2 which is less than the maximum observed at half-monolayer coverages. Here

and in Figs. 2b,3b,4b "monolayers" means the sum of the number of layers deposited plus the number removed. There is of course no change in the average film thickness in cases where growth and sputtering rates are equal.

Moderately rough surfaces

The simple picture of single adatoms and holes annihilating, which obviously leads to smoothening, breaks down once the growth and/or sputtering rates are increased sufficiently to make $\sigma_1^2 > 1$. The situation is more like given in Fig. 2a where one imagines the random growth and sputtering producing regions of excess adatoms or holes. This is the sense of Eq. (2), which shows that unless $\rho(0)$ is near unity for the above very smooth surface, deposition and sputtering make additive contributions to roughness without any cancellation effects. This is confirmed by the simulation results in Fig. 2b for which R_p and R_i have been increased by a factor of 20 over their values in Fig. 1b, all other factors remaining the same. Note that the steady value for either deposition or sputtering alone, $\sigma_1^2 \approx 0.9$, still corresponds to a reasonably smooth surface; but it is sufficiently large to remove any smoothening effects brought about by random creation of holes. The limiting values are in excellent agreement with theory, as shown in refs. 5 and 6.

Inclusion of interactions

Cohesive interactions of the nature discussed above make holes less mobile than adatoms. Thus, as indicated in Fig. 3a, the deficient region produced by sputtering will anneal out more slowly than the excess region produced by deposition; so sputtering leads to rougher surfaces than does deposition. The pertinent simulation data are shown in Fig. 3b, with the interaction parameter $\gamma = 0.9$ and the total rate of deposition plus sputtering the same in all cases.

Binding-dependent sputtering

The same cohesive energy E_c which inhibits breakup of clusters by hopping also should affect the sputtering yield. It is generally accepted [8] that the yield is inversely proportional to E_c for the relatively low ion beam energies used [1-3]. How this might lead to ion-induced smoothening is sketched in Fig. 4a. An ion incident at a site which is not too strongly bound knocks a particle off, which correspondingly makes the particle at a neighbor site free to jump down the step since the cluster has been broken. Thus there is a smoothening effect. If, however, sputtering were completely random as in the above examples, production of a hole at some site on a smooth part of the surface would be equally likely; and, as seen in Figs. 2b and 3b, there is no net smoothening. But if hole production at a fully imbedded (no lateral exposure) site is sufficiently suppressed, net smoothening can result. A form $P_g = 1/(1 + nc)$ where P_g is the probability an incident ion knocks out a particle clustered to nc others ($0 \leq nc \leq 4$ for the square planar lattice) is likely the most palatable simple dependence, but simulations show it not to be strong enough to produce net smoothening, although it does give much less roughening than a P_g independent of nc . Instead simulation data are shown in Fig. 4b for $P_g = 1/(1 + nc)$ for $0 \leq nc \leq 3$, but $P_g = 0$ for $nc = 4$ when the particle is

imbedded with no lateral exposure. Other parameters are the same as in Fig. 3b. (The value of R_i when the ion beam is turned on at about 55 monolayers is the rate of actual knockouts rather than the rate of ion incidence) Ion-induced smoothening is evident. An exponential dependence $P_s = e^{-\alpha n c}$ gave even greater smoothening in a simulation (not shown) with $\alpha = 2.3$. Such dependence might be expected if the ion beam produced thermal desorption from local hot spots, but this seems unlikely [8].

CONCLUSIONS

Except for very low deposition and ion-bombardment rates, where the picture of independent mobile adatoms and holes is valid, random ion-induced sputtering only adds to the surface roughness. If adatoms are noninteracting, a given rate of random sputtering produces the same roughness as an equal rate of random particle deposition. But cohesive interactions make holes less mobile than adatoms (at least for the simple model of cluster interactions presented here), thus causing sputtering to produce rougher surfaces than particle deposition. If the sputtering probability is strongly dependent on binding energy and configuration, it is possible to have ion-induced smoothening. That is, combined particle deposition and ion bombardment produce a smoother surface than from deposition alone. However, it seems necessary to have a dependence stronger than $1/(1 + nc)$, as expected for the simplest models of sputtering, where nc is the number of neighbors on the surface to which a particle is laterally bonded. Whether sputtering yield from a fully bonded surface atom ($nc = 4$) is sufficiently less than that from an isolated adatom ($nc=0$) to produce the desired effects may be an open question worthy of investigation.

The only effect of ion bombardment considered here was sputtering. Ion-induced hopping and creation of adatom-vacancy complexes was neglected. Inclusion of the former mechanism together with a $1/(1 + nc)$ sputtering probability could significantly enhance the tendency toward smoothening.

Work performed at Sandia National Laboratories supported by the US Department of Energy under Contract No. DE-AC04-76DP00789

REFERENCES

1. E. Chason, K. M. Horn, J. Y. Tsao and S. T. Picraux, Mater. Res. Soc. Symp. Proc. 128, 35 (1989); E. Chason, P. Bedrossian, K. M. Horn, J. Y. Tsao, and S. T. Picraux, Appl. Phys. Lett. 57, 1793 (1990).
2. C.-H. Choi, L. Hultman, and S. A. Barnett, J. Vac. Technol. A8, 1587 (1990).
3. E. Chason, J. Y. Tsao, K. M. Horn, S. T. Picraux and H. A. Atwater, J. Vac. Sci. Technol. A8, 2507 (1990).
4. S. B. Ogale, A. Madhukar, and M. Thomsen, Appl. Phys. Lett. 51, 837 (1987).

5. P. M. Richards, Phys. Rev. B, in press.
6. P. M. Richards, Surf. Sci., in press.
7. P. M. Richards, submitted to Phys. Rev. B.
8. P. Sigmund and C. Claussen, J. Appl. Phys. 52, 990 (1981).

Ion smoothening by adatom-hole annihilation

Fig. 1a. Ion-induced smoothening for very smooth surface. Dashed circle represents "hole" produced by sputtering. Arrow shows transport plus hop into hole.

Fig. 1b. Simulated ion-induced smoothening for very smooth surface.

Equal effects of deposition and sputtering

Fig. 2a. Schematic for moderately rough surface. Arrows show hops.

Fig. 2. Lack of ion-smoothing for moderately rough surface.

Slower annealing of ion damage

Fig. 3a. Same as Fig. 2a, but with interactions included to inhibit hop into hole, as indicated by weak arrow.

Fig. 3b. Effect of interactions on difference between growth and sputtering.

Ion smoothening by selective sputtering

Fig. 4a. Schematic with sputtering probability dependent on interactions. Knockout with bold arrow enhances interacting jump rate since it breaks up cluster.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Fig. 4b. Ion-induced smoothening for interaction-dependent sputtering probability.

END

DATE FILMED

12 / 17 / 90