

Grandjean and Massies Reply: In a recent Letter [1], we have shown experimentally that a preadsorbed Te monolayer on a GaAs (001) surface has a strong effect on the subsequent epitaxial growth of InAs which is severely strained (7.2% lattice mismatch). A significant continuation of the pseudomorphic growth regime was observed as a consequence of the suppression of island formation by which strain relaxation occurs in the early stages of growth. Another experimental observation, which is central to the understanding of island inhibition, is that the Te layer is continuously segregated at the growing surface. We interpret such a surface segregation as mainly due to the surface free energy minimization, i.e., Te acts as a surfactant [2]. In order to allow Te atoms to occupy the surface sites, In and As atoms are rapidly incorporated into subsurface sites. As a consequence the In surface diffusion is greatly reduced and 3D islands cannot be formed.

In the preceding Comment [3], Snyder and Orr point out that lowering the surface free energy actually facilitates island formation. This is obviously true, but only for equilibrium or near equilibrium situations where sufficient mass transport is provided by atom surface migration, which is not the case for the surfactant mediated growth. They also state that "the success of surfactants is due to altering kinetics not energetics." We think, however, that kinetics are modified because of energetic change induced by the surfactant. Under usual molecular beam epitaxy growth conditions as we used for this investigation, we can neglect the evaporation rate, i.e., the residence time of impinging atoms is simply the time to grow one monolayer. Thus, at constant growth rate, the surface diffusion length depends only on the diffusion coefficient

$$D = D_0 \exp(-E/kT),$$

where E is the energy barrier to migration which depends on the configuration of the initial site, and k is Boltzmann's constant. When using a surfactant, we should add an energetic barrier which corresponds to the exchange between atoms of the growing film and the surfactant layer imposed by the segregation process. The mean exchange energy per atom E_{ex} can be simply related to the difference between the surface free energies of

the epitaxial layer γ_{epi} and of the surfactant layer γ_{surf} :

$$E_{ex} = (\gamma_{epi} - \gamma_{surf})\sigma,$$

where σ is the surface area per atom. This equation implies that if $\gamma_{surf} < \gamma_{epi}$ then $E_{ex} > 0$ and $D = D_0 \times \exp[-(E + E_{ex})/kT]$ is decreased. Another objection of Snyder and Orr is, however, that macroscopic terms such as the surface free energy may not be related to exchange and incorporation which is a local process. According to these authors, the important feature is the energy difference between local atomic bonding configurations of the surfactant and epilayer. But can we infer that on a macroscopic scale the result will be opposite of a local process? We still think that the variation of macroscopic terms such as the surface free energy reflects local processes. (Note that by definition, the surface free energy is directly related to the atomic bonding configuration at the surface.)

In conclusion, we think that the surfactant term is justified in the present work as well as in previous works by Copel *et al.* [2] and Tromp and Reuter [4]. However, we agree with Snyder and Orr that there is still a great deal of uncertainty in the mechanism of surfactant action.

We thank M. Leroux for fruitful discussions. This work is supported in part by the Centre National d'Etudes des Télécommunications.

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Received 12 October 1992

PACS numbers: 68.55.Bd, 61.14.Hg, 68.35.Bs

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