Extending the epitaxial thickness limit in low-substrate-temperature-grown GaAs

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A method for extending the epitaxial thickness limit in low-temperature-grown GaAs (LT-GaAs) is presented. It is shown that the use of vicinal GaAs(001) substrates with a high misorientation angle reduces the surface roughness of LT-GaAs and inhibits the nucleation of defects which cause the breakdown of perfect epitaxial growth. Kinetic Monte Carlo simulations are used to describe the influence of the vicinal substrate on the growth mode and to estimate the appropriate misorientation angle. © *2001 American Institute of Physics.* [DOI: [10.1063/1.1420783]

Epitaxial growth at low substrate temperatures has been extensively studied in a number of systems such as metals and semiconductors.[1] Apart from the purely scientific interest in understanding the mechanisms of epitaxy, this subject has also great technological importance, mainly in the case of semiconductors. The use of low substrate temperatures during growth of semiconductor heterostructures may reduce unwanted effects like dopant segregation and interface interdiffusion. Furthermore, surface roughening due to strain is depressed, leading to smoother surfaces and interfaces. Especially for GaAs, the growth at low substrate temperatures is most interesting, due to the unique properties acquired by the material, such as ultrashort carrier lifetimes, which make it ideal for fast optoelectronic applications.[2]

One of the problems inherent to low-temperature epitaxy is the existence of a limiting film thickness h_e, beyond which the epitaxial growth turns to polycrystalline or amorphous.[3] h_e is strongly growth temperature dependent, becoming smaller with decreasing substrate temperature T_s. This restricts the range of layer thicknesses that may be employed at a given temperature. For GaAs, values of h_e in the range between 600 nm and 3 μm have been reported for $T_s = 200\,°\mathrm{C}$.[4] The large spread in the reported values is a consequence of the difficulties in accurately determining the on-wafer temperature. As shown recently,[5] this can be avoided by the use of special *in situ* characterization techniques which directly probe the properties of LT-GaAs. There have been many contradicting reports concerning the origin of epitaxy breakdown in GaAs. Some studies suggested that strain plays the central role in the effect.[4,6] Their arguments are based on the well-known crystal expansion of low-temperature-grown GaAs (LT-GaAs) due to excess As incorporation, and on the fact that it grows pseudomorphically strained on the GaAs(001) substrate.[2] Another group proposed that the main reason for the effect is surface roughening,[7] which is enhanced at low growth temperatures due to kinetic effects. Further support for this argument is given by a recent report,[8] showing that the surface morphology of LT-GaAs is characterized by large three-dimensional (3D) growth mounds, which significantly increase the surface roughness. Mound formation in LT-GaAs is attributed to enhanced adatom surface diffusion due to an As self-surfactant layer.[8] Nevertheless, there is no conclusive evidence so far justifying either of the two proposed mechanisms of epitaxy breakdown.

In this letter, we show that there is a way to push the h_e limit to higher values for LT-GaAs. If we assume that surface roughness causes the breakdown of perfect epitaxial growth, then it should be possible to avoid this breakdown by making the surface smoother. A technique to accomplish this in epitaxial systems exhibiting mound formation indeed exists, and it was proposed by Johnson *et al.*[9] in their pioneering article on surface mound formation. According to their work, growth on a *vicinal* substrate suppresses the formation of growth mounds and, thus, produces smoother surfaces. As we will demonstrate, this idea works also for LT-GaAs, because its surface roughness originates from mound formation as well.[8] Our results support the idea that surface roughness is the origin for the breakdown of epitaxy in LT-GaAs.

The key question arising is, what should be the angle ϕ and direction of substrate misorientation in the case of LT-GaAs. To answer this, we have to consider in some detail the mechanisms of mound formation. It is by now generally accepted that mounds occur due to the Ehrlich–Schwoebel (ES) surface diffusion barrier, which hinders the down movement of adatoms at surface step edges.[9] As a result, adatoms tend to accumulate on top of already existing growth islands, and so finally the large 3D growth structures called mounds occur. The parameter which governs the development of mounds on a vicinal surface is the ratio of the average separation between two-dimensional islands during deposition of the first layer, σ, to the length of terraces, l; if $l < \sigma$ mound formation will be suppressed.[9] Taking into account the estimation for the surface hopping barrier $E_a \sim 0.5$ eV reported in Ref. 8 for LT-GaAs, and the relation $\sigma \approx a(H/R)^{1/6}$,[10] where a is the surface lattice constant, $H \cong (k_B T/h)\exp(-E_a/k_B T)$ the hopping rate, and R the growth

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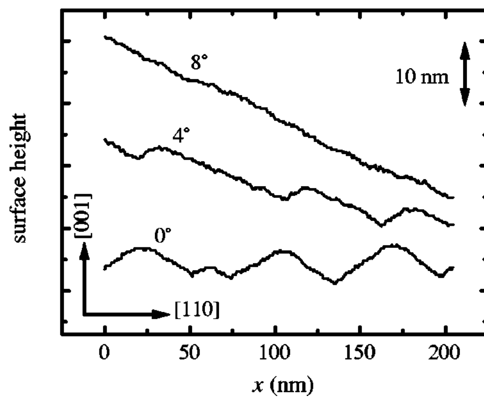

FIG. 1. Surface profiles obtained from numerical simulations of epitaxial growth on singular and vicinal substrates. The respective misorientation angle of the substrate towards (111)A is given on each curve. Curves are shifted vertically for clarity.

rate, we can estimate σ to be of the order of $18a$ at $T_s = 200\,°\mathrm{C}$ and $R = 1$ monolayer/s. This means that $\phi > 2°$ has to be satisfied in order for mound formation to be suppressed, since $\tan\phi = a/\sqrt{2}l$. Furthermore, since mounds on LT-GaAs are elongated along $[\bar{1}10]$ and the larger undulations of the surface occur along [110],[8] the misorientation has to be towards [110] or $[\bar{1}\bar{1}0]$ [towards the $(111)A$ planes], in order to be most effective in suppressing the surface roughness.

To test these assumptions and to improve on the aforementioned crude estimate of ϕ, we also performed kinetic Monte Carlo simulations of the epitaxial growth for several misorientation angles. Our model is based on Ref. 11; it considers random deposition of atoms on the (001) surface of a face-centered-cubic lattice and subsequent thermally activated surface diffusion at a rate $H \sim \exp(-E_a/k_BT)$. The rate for hopping down a step edge is $H' = H\exp(-E_s/k_BT)$, where E_s is the ES barrier. Details of the simulation procedure will be published elsewhere. We chose the values $E_a = 0.7\,\mathrm{eV}$ and $E_s = 0.1\,\mathrm{eV}$, which closely reproduce the surface morphology of LT-GaAs grown on singular substrates. Surface profiles obtained from simulating the growth of 1 μm thick layers on substrates of different misorientations are depicted in Fig. 1. Mound formation is clearly evident for $\phi = 0°$, i.e., for a singular substrate. As ϕ increases, mound formation is suppressed. At $\phi = 8°$, mounds disappear completely and the surface profile is smooth. However, the use of such a high misorientation imposes some technical problems; for instance, it is difficult to prepare a high quality starting surface due to step-bunching instabilities. Hence, in our experiments, we utilize a 4° vicinal substrate, which, according to the simulation, should significantly reduce the surface roughness, and at the same time can be readily handled.

LT-GaAs samples were grown by solid-source molecular-beam epitaxy (MBE) on nominally singular and vicinal (4°→(111)A) GaAs(001) wafers. A 250 nm thick GaAs buffer layer was grown at 580 °C, to establish a smooth and clean starting surface. Low temperature growth was conducted at a substrate temperature of 200 °C, with a growth rate of 1 μm/h and an As_4:Ga beam equivalent pressure ratio of 30.

The lattice expansion of LT-GaAs epilayers with respect to the substrate was examined by means of double crystal x-ray diffraction. Rocking curves around the (004) reflection are shown in Fig. 2 for two 850 nm thick layers grown on a singular and a vicinal substrate, respectively. For the singular sample, the substrate-epilayer peak separation corresponds to the lattice mismatch $(\Delta a/a)_\perp$ in the growth direction, which is estimated to be 0.14%. The situation is different for the misoriented sample. The two peaks labeled (a) and (a′) correspond to scans at two different azimuths: one parallel to the direction of misorientation and one 180° apart. The separation between them, 40 arcs, is equal to the angle between the (001) planes in the epilayer and substrate.[12] $(\Delta a/a)_\perp$ can be found from the mean value of the separation of peaks (a) and (a′) from the substrate peak respectively.[12] It is easily seen that $(\Delta a/a)_\perp$ of the misoriented sample coincides with that of the singular one, hence demonstrating identical growth conditions. The only difference is that the peak from the singular sample appears weaker and broader, indicating an inferior crystal quality of this epilayer.

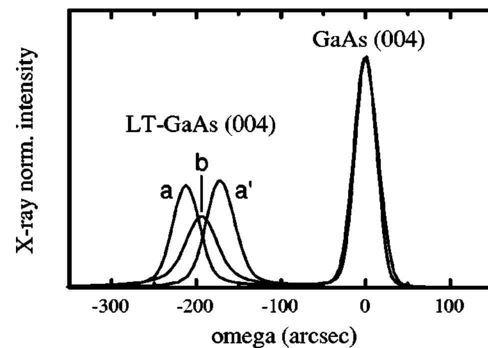

FIG. 2. X-ray diffraction rocking curves from LT-GaAs layers grown at 200 °C on vicinal (curves a and a′) and singular (curve b) GaAs(001) substrates.

This finding was confirmed by transmission electron microscopy (TEM). A cross sectional bright-field TEM image of the layer grown on the singular GaAs(001) substrate is shown in Fig. 3(a). The clearly distinguished pyramidal-shaped defects are typical for LT-GaAs. They are known to initiate the deviation from perfect epitaxial growth, and to gradually drive the material to polycrystallinity.[6] The defect-free layer has a thickness of 650 nm on the average, which corresponds to the limiting epitaxial thickness h_e. This value is in agreement with previous reports for this growth temperature.[4] In contrast, the LT-GaAs layer grown under identical conditions on a vicinal substrate, Fig. 3(b), is completely free of structural defects. We were able to observe only one such defect, which is marked with an arrow in Fig. 3(b), in the approximately 100 μm long cross section of the TEM specimen. Obviously, the value of h_e is larger than the actual film thickness in this case.

The insets of Figs. 3(a) and 3(b) depict the morphology of the surface, as could be imaged by TEM in regions of the specimens where the surface was not damaged by the specimen preparation procedure. The electron beam was parallel to the $[\bar{1}10]$ direction of the GaAs lattice, thus the images display a profile of the surface along [110]. As seen in the inset of Fig. 3(a), the surface of the singular sample shows large pyramid-like undulations, with ~8 nm height and ~50 nm base. It should be noted that measurements were taken at regions with no pyramidal defects beneath the sur-

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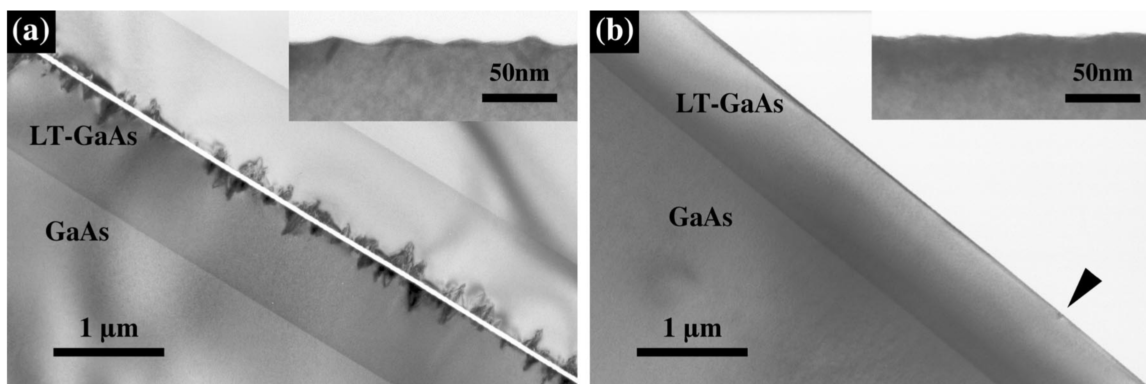

FIG. 3. Cross sectional transmission electron microscopy bright-field images of LT-GaAs layers grown on (a) a nominally singular and (b) a vicinal GaAs(001) 4°→(111)A substrate. The insets depict the surface of the samples in larger magnification.

face. Our observations are in complete accordance with Ref. 8 and with our growth simulations. The undulations seen in the inset of Fig. 3(a) are cross sections of growth mounds, and their dimensions agree very well with those reported in Ref. 8. In the case of the misoriented sample, the surface undulations are observed as well [inset of Fig. 3(b)], but with a much lower height ~ 2 nm. This is again in agreement with the Monte Carlo simulations.

We conclude that our epitaxial model describes well the growth of LT-GaAs on vicinal substrates. As predicted by the simulations and verified experimentally, mound formation is significantly suppressed and surface roughness decreases when LT-GaAs is grown on vicinal substrates. Furthermore, the nucleation of pyramidal defects is delayed, raising the epitaxial thickness limit h_e to higher values. Our results show that surface roughness, in the form of mounds, plays the key role in the breakdown of epitaxial growth, since our investigations are performed on samples with the same amount of strain. However, there is still no evidence on the exact mechanism causing the nucleation of pyramidal defects. There is no doubt though, that the growth mounds which characterize the surface of LT-GaAs, their dimensions, and shape, definitely have a relation to the pyramidal defects. Further reserach is required, in order to clarify this relation.

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