Suppression of Three-Dimensional Island Nucleation during GaAs Growth on Si(100)

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Very-low-energy (≈ 28 eV), high-flux (≈ 0.4 mA/cm^2) Ar-ion irradiation during molecular-beam epitaxy changed the nucleation of GaAs on Si(100) from Stranski-Krastanov to a mechanism approaching layer-by-layer growth. While three-dimensional island nucleation was eliminated, the growth surface exhibited low-amplitude undulations. The results are explained by ion-induced removal of atoms from stable 3D islands, which suppressed 3D island nucleation.

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Heteroepitaxial film growth onto substrates with much different lattice constants, crystal structures, and/or chemistry generally results in the nucleation of three-dimensional (3D) islands. Examples of lattice-matched heterostructures where 3D island nucleation occurs include GaP/Si [1], GaAs/Ge [2], and Si/CaF_2 [3]. Studies on chemically and structurally similar alloy systems such as InGaAs/GaAs [4], InGaAs/InP [5], InAlAs/InP [5], and GeSi/Si [6] show that Stranski-Krastanov (SK) nucleation occurs for sufficiently large mismatch.

3D island nucleation during epitaxy is undesirable for two reasons. First, planar device structures involving quantum wells and superlattices require flat interfaces. Second, defects are introduced into heteroepitaxial layers during 3D island growth and coalescence [1,7–9]. There has thus been considerable interest in developing methods for controlling nucleation characteristics. In some cases, reducing the nucleation temperature suppresses 3D island nucleation during SK growth [10]. Adsorbing a foreign surface species (e.g., Sb or As) has recently been shown to enhance layer-by-layer growth of Ge on Si and vice versa [11]. In other cases, however, changes in growth conditions are not successful in eliminating 3D islands. GaAs on Si(100) grown by molecular-beam epitaxy (MBE) is a good example [12]. Attempts to modify the nucleation mechanism by varying the nucleation temperature [13,14] and reducing the As overpressure [15] increase the saturation 3D island density and hasten film coalescence. While this yields flat surfaces earlier during growth, 3D island nucleation is not eliminated. The use of migration-enhanced epitaxy also does not eliminate 3D island nucleation [16].

In this Letter, we describe the use of ion-assisted MBE (IAMBE) to control the nucleation of GaAs on Si. A similar ion-assisted deposition technique was previously used to delay the nucleation of 3D InAs islands on Si(100) from ≈ 1 monolayer (ML) to ≈ 10 ML [17,18]. The present results for GaAs on Si(100) show that very-low-energy (~ 30 eV), high-flux (~ 0.4 mA/cm^2) Ar-ion irradiation changed the nucleation characteristics from SK to a mechanism approaching layer-by-layer growth; i.e., 3D island nucleation was eliminated.

The IAMBE system was described in detail previously [19]. Briefly, the ultrahigh vacuum system is similar to conventional MBE chambers, with four effusion cells, a computer-controlled shuttering system, liquid-nitrogen-cooled shrouds, a sample-insertion load lock, and a reflection high-energy electron-diffraction (RHEED) system. The unique feature of the system is the triode discharge that provides high-flux (0.03–1.0 mA/cm^2), very-low-energy (10–200 eV) Ar ions at the film surface during deposition. The Ar-ion energy E was determined from the applied substrate bias, measured relative to the discharge anode, based on plasma potential measurements [17].

The polished (100) p-type Si (20–40 Ω cm) substrates were cleaned using organic solvents before insertion into the growth chamber. Prior to deposition, substrates were outgassed at 600 °C for 5 min, sputter cleaned for 15 min, and then annealed at 800 °C for 2 min. Sputter cleaning was performed with 100-eV Ar ions at a flux $J=0.4$ mA/cm^2 and substrate temperature $T_s=380$ °C. RHEED patterns observed after this procedure were streaky with second-order reflections, indicating that the Si surface was flat and (2×1) reconstructed.

MBE and IAMBE experiments were carried out under identical conditions except for the ion irradiation during IAMBE. Since the IAMBE experiments required an Ar pressure of 2.5 mTorr to operate the triode discharge, both IAMBE and MBE were carried out at this pressure to ensure that a change in gas ambient did not influence nucleation. However, the only effect observed due to the Ar ambient was scattering of the molecular beams, which caused an $\sim 80\%$ reduction in the growth rate, to 0.15 μm/h, at an 18-cm source-substrate separation. RHEED patterns were taken immediately after growth was completed and the Ar gas evacuated. In the results below, the GaAs surfaces were stable during RHEED observations.

Figure 1 shows RHEED patterns taken from GaAs grown on Si at $T_s=380$ °C to different nominal thicknesses using MBE and IAMBE. The As_4/Ga flux ratio was ≈ 2. $E=28$ eV and $J=0.4$ mA/cm^2 were used during IAMBE. In the MBE case, the streaky RHEED pattern observed at low coverage changed to a bulk spot pattern after ≈ 2 ML of deposition, indicating that 3D GaAs islands had nucleated. This result is in agreement with previous reports showing SK growth with a critical

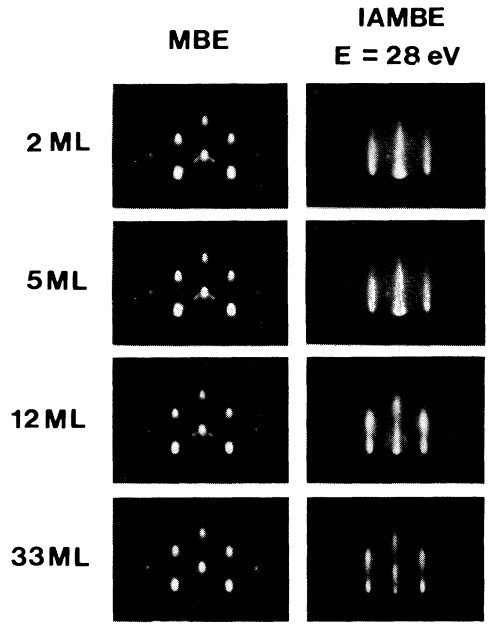

FIG. 1. RHEED patterns from GaAs films grown using MBE and IAMBE to nominal coverages ranging from 2 to 33 ML. $E=28$ eV and $J=0.4$ mA/cm^2 were used during IAMBE.

thickness for 3D island formation of 2–4 ML [12]. The weak spots observed between the fundamental diffraction spots were due to twins. The RHEED pattern became streaky after ~ 100 nm of deposition, suggesting that 3D islands had coalesced and that the growth surface had smoothened. In the IAMBE case, a streaky RHEED pattern was observed at all thicknesses, showing that 3D islands were not present. However, streak intensity modulations, with maximum intensities at the bulk spot positions, developed at thicknesses >5 ML. This indicated that the surface was undulating. Continued growth to thicknesses >33 ML resulted in sharper streaks, decreased intensity modulation along the streaks, and the appearance of higher-order streaks.

Figure 2 shows cross-sectional transmission-electron-microscope (XTEM) images from 18-ML-thick GaAs films grown on Si by MBE and IAMBE, using the same conditions as in Fig. 1. In the MBE case [Fig. 2(a)], 3D GaAs islands were observed, in agreement with previous reports [13]. After IAMBE, a continuous GaAs layer was observed [Fig. 2(b)]. XTEM observations over a range of film thicknesses confirmed that 3D islands were not present under these irradiation conditions, in agreement with the RHEED results. The lack of a well-defined GaAs free surface in Fig. 2(b), combined with the intensity modulation along the RHEED streaks shown in Fig. 1, indicates that the GaAs surface exhibited slight undulations. While it is not clear in the micrographs shown, higher magnification images show that both MBE and IAMBE films exhibited lattice fringes. This, combined with the RHEED patterns which were similar to the 12-ML patterns in Fig. 1, shows that the films were epitaxial. In addition, when 0.5-μm-thick GaAs films were grown by MBE on 60-nm-thick MBE and IAMBE buffer layers, the x-ray rocking curve full width at half maximum was 1340 arc sec for the film with the IAMBE buffer layer, versus 1700 arc sec for the MBE film. This suggests that the crystalline perfection of the buffer layers was improved by ion irradiation.

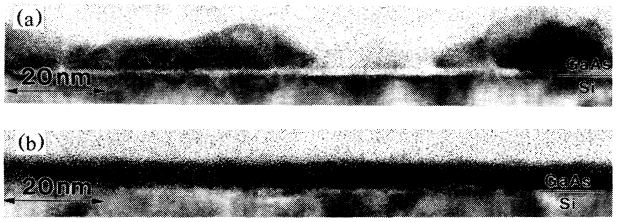

FIG. 2. XTEM micrographs of nominally 18-ML-thick GaAs films grown using (a) MBE and (b) IAMBE. $E=28$ eV and $J=0.4$ mA/cm^2 were used during IAMBE. Note that the contrast above the film surface was from amorphous carbon deposited to protect the surface during XTEM sample preparation.

Many of the ion-surface interaction effects that are commonly used to explain ion-induced changes in film growth [20] cannot explain the results shown above. Ion-enhanced adatom surface diffusivities, often used to explain changes in 3D island densities during nucleation, would tend to enhance the rate of 3D island growth in the present case. Furthermore, for the very low energies used in the present experiments, effects such as preferential sputtering and ion mixing [21] of the GaAs/Si interface are expected to be negligible.

Three mechanisms can be proposed to explain the observed ion-assisted nucleation effect. First, ion-induced removal of atoms can render clusters thermodynamically unstable by either changing their geometry or reducing the number of atoms below the critical size for stability [22]. Second, forward sputtering of material from the sides of 3D surface features onto the underlying substrate can smooth surfaces [23]. Third, ion irradiation might change the rate of relaxation of the 4% lattice mismatch by creating defect sites for nucleation of dislocation half loops. Based on the theory of SK nucleation proposed by Price [24] and Stoyanov [25], this would be expected to change the nucleation characteristics.

In order to give a clearer picture of the ion-assisted nucleation mechanism, experiments in which both the growth fluxes and ion irradiation were pulsed, rather than continuous, were carried out. In these experiments, $E=33$ eV and $J=0.2$ mA/cm^2 were used. First, a film was grown for 120 sec at $T_s=380\,^\circ$C by MBE, yielding a nominally 18-ML-thick film, and then bombarded by Ar ions for 120 sec. The spotty RHEED pattern became only slightly more streaky after irradiation, in contrast to

the streaky patterns obtained when growth and irradiation were simultaneous. Second, a migration-enhanced epitaxy cycle was utilized. The Ga and As fluxes were alternated at intervals of 8 sec each, without pauses, yielding $\simeq 1$ ML of GaAs per cycle. Figure 3 shows the RHEED patterns taken from GaAs grown at $T_s = 380\,^{\circ}\mathrm{C}$ after 70 ML of deposition. Migration-enhanced epitaxy without ion irradiation yielded a spotty RHEED pattern [Fig. 3(a)] similar to that observed for MBE. When ion irradiation was carried out during both the Ga and As exposures [Fig. 3(b)], or during the Ga exposures only [Fig. 3(c)], RHEED patterns similar to the IAMBE case were observed. However, when ion irradiation was used only during the As exposure, a RHEED pattern [Fig. 3(d)] only slightly more streaky than the MBE case was obtained.

These results show that 3D island nucleation was only suppressed when ion irradiation was concurrent with Ga deposition. This is consistent with the idea that 3D GaAs islands nucleate due to the aggregation of Ga adatoms [13]. Regarding the mechanisms proposed above, the following conclusions can be made. Smoothing by forward sputtering and redeposition did not play the key role since there was little effect when the surface was already covered with stable 3D islands. Irradiation effects on lattice relaxation also did not play a major role, since any effect should occur during either the Ga or As deposition.

These observations suggest that ion irradiation intervened directly in the nucleation of 3D islands. The rate of formation, dn_s/dt, of stable 3D nuclei can be written in terms of the rate of diffusion of single adatoms, with surface density n_1 and surface diffusion coefficient D, to critical-sized clusters with surface density n_{cr} [10]. The effect of ion-induced displacements on the nucleation rate can be estimated by adding a term for the breakup of stable nuclei:

$$dn_s/dt \approx \sigma D n_1 n_{cr} - p(E) J A n_{cr+1} . \quad (1)$$

The first term on the right-hand side is the usual nucleation rate expression, where σ is a geometrical factor describing adatom diffusion to islands, typically 2–4 [10]. The second term is the rate at which barely stable nuclei with surface density n_{cr+1} are rendered unstable by ion-induced removal of an atom. $p(E)$ is the average number of displacements per ion with energy E and A is the average surface area covered by a stable nucleus.

Equation (1) can be evaluated by making the following simplification. The surface densities n_j of clusters with j atoms are given by the Walton relation [26]

$$n_j/N = (n_1/N)^j [\exp(-\Delta G_j/kT_s)] , \quad (2)$$

where N is the surface atomic density, G_j is the free-energy change when j adatoms combine to form a cluster, and k is the Boltzmann constant. Taking $\Delta G_{cr} \approx \Delta G_{cr+1}$ since the cluster free energy versus j is at a maximum at $j = j_{cr}$ [10], Eq. (2) gives $n_{cr+1}/n_{cr} \approx n_1/N$. Equation (1) can thus be rewritten as

$$dn_s/dt \approx [\sigma D - p(E) J A/N] n_1 n_{cr} . \quad (3)$$

When the rate of ion-induced removal of atoms from j_{cr+1} clusters exceeds the rate of adatom addition to j_{cr} clusters, i.e., when $p(E)JA/N > \sigma D$, 3D nucleation is suppressed. Initial investigations of the J dependence showed that 3D island nucleation was suppressed for $J > 0.2$ mA/cm^2 with $E = 28$ eV. Using $p(E = 30\ \mathrm{eV}) \approx 0.2$ [17], $N = 6.5 \times 10^{14}$ cm^{-2}, estimating $A \sim 10^{-14}$ cm^{-2} from $j_{cr} \sim 10$ [10], and extrapolating $D \approx 10^{-15}$ cm^2/sec for GaAs at 380 °C [27] gives $p(E)JA/N > \sigma D$ when $J > 0.1$ mA/cm^2, in good agreement with experiment considering the approximate nature of the input values. Initial experiments on the effect of E were also carried out. $E < 20$ eV had little effect on nucleation; these energies were too low to displace atoms [17] leading to $p \sim 0$ in Eq. (3).

The above results are the first indication, to our knowledge, that GaAs can be grown on Si(100) without 3D island nucleation. Previous results on the ion-assisted deposition of InAs on Si showed that 3D island formation was suppressed during the first ~ 10 ML of deposition, but that 3D islands invariably nucleated at higher thicknesses [17,18]. The difference between this and the present result was presumably due to a larger driving force for 3D island nucleation for InAs on Si, as a result of its larger lattice mismatch (11% vs 4% for GaAs/Si).

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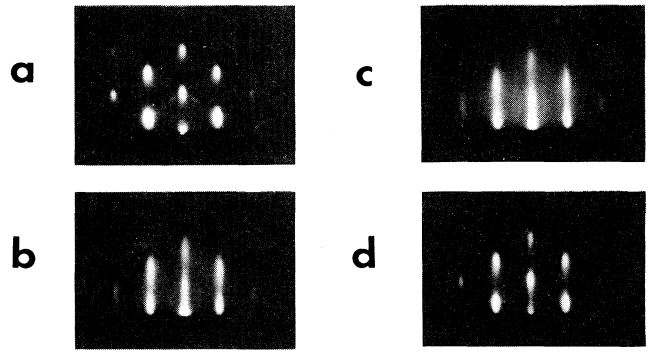

FIG. 3. RHEED patterns observed from 70-ML-thick GaAs films grown using migration-enhanced epitaxy with (a) no ion irradiation, (b) irradiation during both the Ga and As flux exposures, (c) irradiation during the Ga flux exposures, and (d) irradiation during the As flux exposures. $E = 33$ eV and $J = 0.2$ mA/cm^2 during the irradiations.

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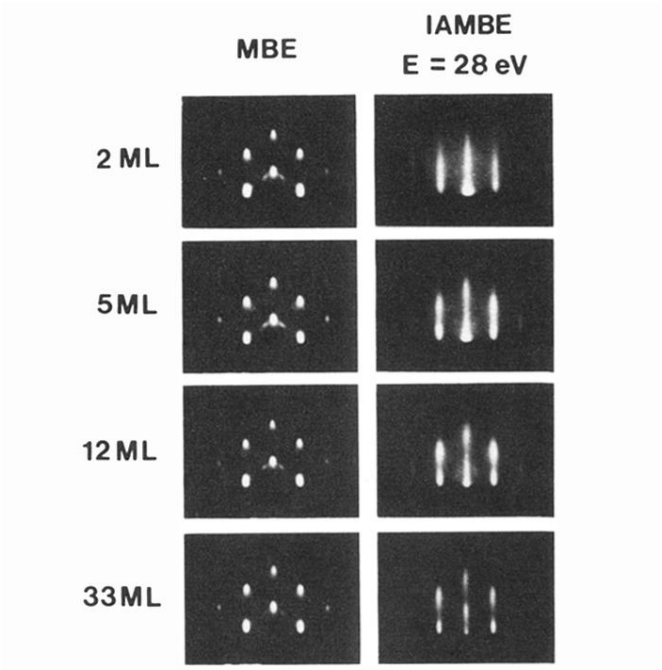

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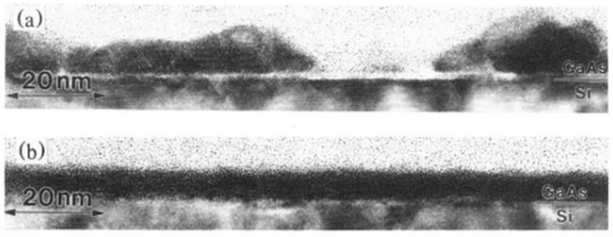

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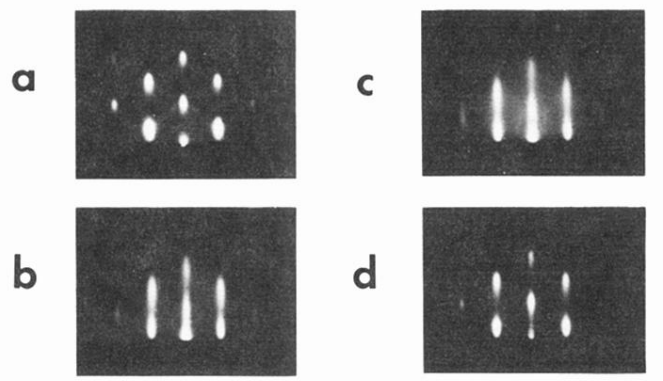

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