Glancing-Angle Ion Enhanced Surface Diffusion on GaAs(001) during Molecular Beam Epitaxy

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We describe the effects of glancing incidence 3–4 keV Ar ion bombardment on homoepitaxial growth on vicinal GaAs(001). The average adatom lifetime on surface terraces, measured during growth using specular ion scattering, decreased monotonically with increasing ion current density. The results indicated that surface diffusivity was increased by the ions. The ion beam also suppressed growth oscillations and decreased the film surface roughness. This indicates a change from two-dimensional island nucleation to step-flow growth due to increased adatom surface diffusivity. A simple model, involving direct momentum transfer from ions to adatoms, is shown to be consistent with the measured enhanced diffusion.

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Ion beam assisted deposition (IBAD) is an important technique for modifying the structure and properties of thin film materials [1–3]. One feature of IBAD that has been widely discussed is enhanced surface diffusion. Much of the evidence for this is indirect, e.g., reductions in minimum temperature for epitaxial growth [2] or changes in island size distributions during Volmer-Weber nucleation [1]. Recently, Ditchfield and Seebauer directly observed Ar ion-enhanced Ge surface transport on Si(111) using optical second harmonic microscopy [4]. Simulations of ion-surface interactions during film deposition indicate that a number of near-surface atoms are displaced during a typical ion impact [5].

Much of the above work utilized ions impinging at angles between 0° and 60° from the surface normal. Studies of glancing-incidence angle ions, defined as angles $\phi \approx 1°–10°$ relative to the surface plane, have been confined to modifications of static surfaces rather than growth surfaces [6]. However, these studies suggest unique possibilities for film growth. In particular, glancing-incidence ions are mostly reflected rather than penetrating at flat defect-free portions of a surface, resulting in minimal ion damage, gas entrapment, or sputtering [6,7]. On the other hand, the ions can transfer considerable momentum to surface defects. The momentum transfer to surface adatoms would be mainly in the film plane, a condition that should be ideal for promoting lateral surface diffusion. Thus, glancing-angle ions have the potential to selectively couple momentum to the surface defects, i.e., adatoms and step edges, that control film growth.

In this Letter, we describe the effects of glancing-angle ions on GaAs(001) homoepitaxy. The experiments were performed in a molecular beam epitaxy (MBE) chamber using Ga and As_4 beams. The substrates were semi-insulating, double-polished vicinal GaAs(001) wafers misoriented by 2.5° ± 0.5° towards $(1\bar{1}0)$. The chamber contained a specular ion current measurement (SICM) system that has been described elsewhere [8–11]. A 3–4 keV Ar ion beam, incident at $\phi = 3°$ relative to the nominal substrate surface plane, was used both for monitoring and modifying growth. The scattered ion beam, detected using a movable Faraday cup, was used to measure surface diffusion and growth oscillations. While glancing-angle ions are specularly reflected from locally flat portions of the surface, ions incident near surface defects are nonspecularly scattered, decreasing the specular current. That is, changes in the specular current can be related to changes in the adatom density and step edge length.

In order to characterize adatom diffusion, SICM measurements were performed under growth conditions where the density of 2D islands nucleating was low, i.e., adatom scattering dominated [9]. This was achieved by depositing small coverages [<0.1 ML (monolayer) of GaAs] using low fluxes ($J_{Ga} = 0.1$ ML/sec) on vicinal GaAs (miscut = 2.5°). Figure 1 shows typical SICM results taken at substrate temperature $T_S = 520\,°C$ using 3 keV Ar ion current densities J_i of 1.4 and 3.1 $\mu A/cm^2$. The results are normalized to the pregrowth specular ion current. The current drops during deposition, as the Ga adatom population n_A increases, and then increases following deposition as adatoms diffuse to the step edges (average separation $x = 6.5$ nm), as given by [9]

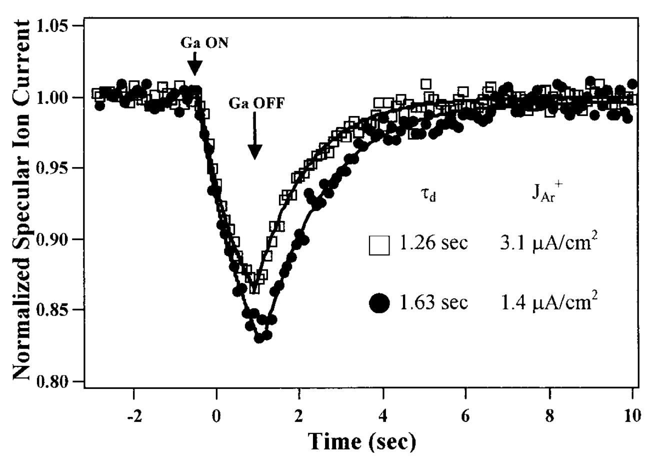

FIG. 1. Specularly scattered ion current vs time before, during, and after GaAs growth at $T_S = 520\,°C$ and 0.1 ML/sec. The deposition time was 1 sec and the ion energy was 3 keV.

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$$\frac{dn_A}{dt} = J_{\mathrm{Ga}} - \frac{n_A}{\tau_d}, \qquad (1)$$

where J_{Ga} is the Ga flux and τ_d is the diffusion time constant. Equation (1) gives simple exponential variations in n_A that fit the data in Fig. 1 well and yield $\tau_d = 1.63$ sec for $J_i = 1.4\ \mu\mathrm{A/cm^2}$, and 1.26 sec for 3.1 $\mu\mathrm{A/cm^2}$. Measured adatom time constants, plotted versus J_i in Fig. 2, gradually decreased with increasing J_i. Values were essentially identical for 3 and 4 keV Ar ions within experimental error. Surface diffusion coefficients can be obtained from this data using $D = x^2/\pi^2\tau_d$ [9].

The decrease in the measured time constant can potentially be explained by direct sputtering of atoms from the surface. However, three independent measurements have shown that sputtering accounted for <10% of the observed changes in τ_d. First, GaAs growth oscillations were measured and the change in oscillation period with J_i was less than the experimental error of 3% [12]. Second, attempts to measure sputtering of an InAs monolayer on GaAs by 1 keV, $\phi = 3°$ Ar ions indicated a very low sputter yield of <0.004 [8]. Third, x-ray diffraction measurements and simulations showed no change in the period of InGaAs/GaAs superlattices due to 3 keV, $\phi = 3°$ Ar ions. The low sputtering yield is due to the low probability of penetration for the glancing incidence ions [6,7].

A simple model was developed assuming enhanced diffusion by direct ion-adatom momentum transfer. The rate of change of the adatom density n_A is given by

$$\frac{dn_A}{dt} = J_{\mathrm{Ga}} - n_A\left[\frac{1}{\tau_d} + J_i\,\frac{\sigma_d}{\overline{v}_I}\right]. \qquad (2)$$

The additional term in Eq. (2), compared with Eq. (1), is the ion effect. σ_d is the ion-adatom interaction cross section, defined by the requirement that the ion transfers sufficient kinetic energy to the Ga adatom to induce a permanent displacement. $\overline{v}_I$ is the average number of ion impacts required to move the adatom to a step edge, i.e., the terrace width divided by the average ion-induced displacement distance. Comparing with Eq. (1), the bracketed term in Eq. (2) can be equated to $1/\tau_m$, where τ_m is the measured J_i-dependent time constant. Rearrangement yields

$$\tau_m = \frac{\tau_d}{1 + J_i\dfrac{\sigma_d}{\overline{v}_I}\tau_d}. \qquad (3)$$

The fit of Eq. (3) to the data in Fig. 2 yielded $\sigma_d/\overline{v}_I = 2.1\ \mathrm{nm^2}$.

To determine if this value was reasonable, we approximated σ_d using the binary collision approximation with a Thomas-Fermi-Moliere potential [13]. We assumed a threshold energy of 2.5 eV or $\frac{1}{4}$ of the bulk displacement energy [14] was required to displace a surface atom from its site. Impact parameters up to ~0.13 nm provided enough energy for adatom displacement. Projecting this circular area along the surface with $\phi = 3°$ yielded $\sigma_d \sim 4\ \mathrm{nm^2}$. Finally, we assume $\overline{v}_I \geq 1$, i.e., that adatoms impacted by energetic Ar ions can move distances up to the terrace width. The displacement of energetic atoms over significant distances while confined within the surface potential was previously predicted by Dodson [15]. The resulting predicted value of $\sigma_d/\overline{v}_I \leq 4\ \mathrm{nm^2}$ is comparable to that obtained from the data in Fig. 2. Thus, the above model, based on the assumption of direct momentum transfer to adatoms, provides a plausible explanation of the enhanced diffusion results.

Changes in surface diffusivities are expected to have significant effects on crystal growth. To further test ion-enhanced diffusion, experiments were carried out on vicinal GaAs, under conditions near the transition between the two-dimensional island nucleation growth mode and step-flow growth, i.e., where the adatom diffusion length is comparable to the vicinal terrace width [16]. The change in growth mode is readily monitored by growth oscillations. In the present experiments, we have used SICM oscillations [11], which are similar to reflection high-energy electron diffraction (RHEED) oscillations [16].

The effect of 4 keV Ar ions on homoepitaxial growth on vicinal GaAs(001) is shown in Fig. 3. The specular ion current oscillations present at $J_i = 0.6\ \mu\mathrm{A/cm^2}$ were suppressed by increasing J_i to 3.1 $\mu\mathrm{A/cm^2}$. This result can be explained by increased diffusivity due to the increased J_i. In addition to enhancing diffusion, ions may also promote step-flow growth by breaking up 2D islands. Note that the percentage ion-current drop was smaller for the higher J_i, indicating a flatter surface. Thus, the loss of oscillations was not readily explained by ion-induced damage or roughening. Chason *et al.* [17] have shown that near-normal incidence ions modify RHEED oscillations by creating surface vacancies that can annihilate with deposited adatoms. However, the vacancy creation is associated with sputtering, and sputter yields are very low for

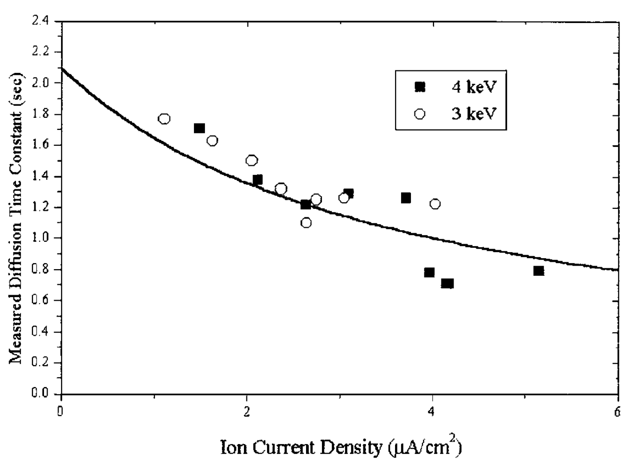

FIG. 2. Surface diffusion time constants obtained from measured specular current variations for 3 and 4 keV Ar ion beams. The curve shown was obtained using Eq. (3).

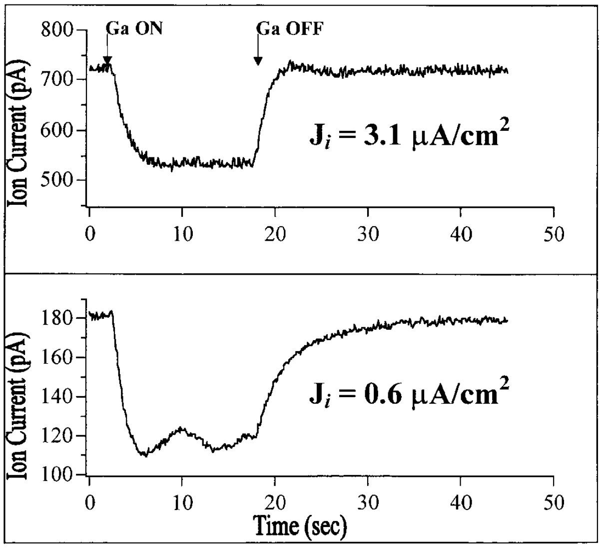

FIG. 3. Specularly scattered 4 keV Ar ion current vs time before, during, and after GaAs(001) growth for two different ion currents. Increasing the current density from 0.6 to 3.1 $\mu A/cm^2$ eliminated the growth oscillations.

small ϕ as discussed above [6,7]. Thus, the ion-induced change in Fig. 3 was not likely to be the result of surface vacancy creation.

If the above interpretation of Fig. 3 is correct, then 2D islands should be present during growth at low J_i, but not at high J_i. Ion-current recoveries after growth have previously been shown to take longer when 2D islands are present [10]. In Fig. 3 and other similar data, the recovery is faster for higher J_i, consistent with an ion-induced transition to step-flow growth.

The effect of ions was further tested by growing 250-nm-thick GaAs films, using the same conditions as in Fig. 3. Atomic force microscopy (AFM) was used to measure surface morphology after growth. Figure 4 shows the surface morphology of the sample grown (a) with and (b) without concurrent 4 keV Ar ion bombardment with an ion current density of 5.2 $\mu A/cm^2$. The nonirradiated MBE film showed an undulating surface with ridges oriented along [110] crystallographic directions with an rms roughness of 0.5 nm. Undulations are commonly observed in MBE films [18]. The morphology during ion-assisted MBE growth was similar, but the magnitude of the rms roughness decreased to 0.25 nm. This decrease is consistent with the idea that ions promoted step-flow growth by enhancing adatom diffusion.

These films were also probed using high-resolution x-ray rocking curves of the (004) GaAs substrate peak to examine the effect of the ions on the crystal quality. Measurements were made using a diffractometer fitted with a tangential drive four-bounce double-channel-cut monochromator. The nonirradiated MBE sample had a full width at half maximum (FWHM) of 22 arc sec, as expected for high-quality homoepitaxy. The sample grown with concurrent ion bombardment had a substantially larger FWHM of 220 arc sec, indicating ion damage. However, the amount of damage was surprisingly small for 4 keV Ar ions at a current density that corresponded to an ion-to-deposited atom ratio of 0.25. Using the standard formula [19] for estimating the number of displaced atoms caused by an ion, $N \approx 0.5E_i/E_D$, where E_D is the displacement threshold for Ga and As atoms in GaAs (9–10 eV) [14], yields ≈50 displacements per ion. This should be enough to severely damage a crystal; however, in the present case the glancing-angle ions are mostly reflected without penetrating the surface, yielding relatively little damage.

The above results suggest that glancing-angle ion momentum can be coupled selectively to adatoms, thereby

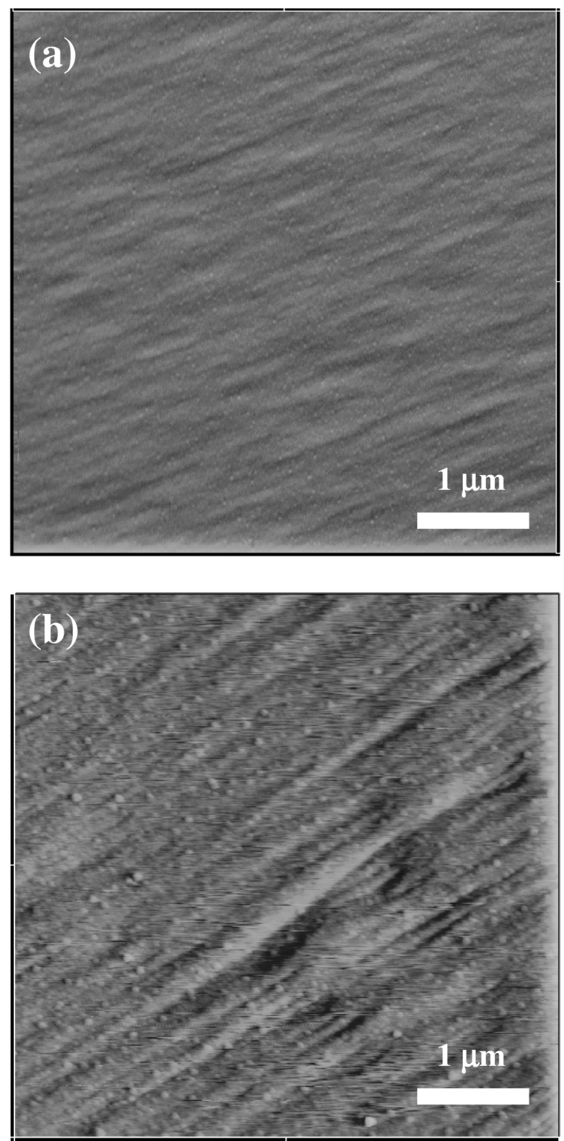

FIG. 4. Atomic force microscope images of the epitaxial GaAs(001) films grown (a) with and (b) without concurrent glancing-angle 4 keV Ar bombardment.

enhancing surface diffusion with relatively minor ion damage. While this technique will likely have little technological application for damage-sensitive semiconductors, there may be important uses for low-temperature growth of more damage-tolerant materials. Examples include oxide superconductors and ferroelectrics, where low temperatures would be useful to minimize interdiffusion and reactions with the substrate.

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